FLINT-CLAY FACIES ILLUSTRATED WITHIN ONE DEPOSIT OF REFRACTORY CLAY

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Abstract--The flint-clay facies, originally proposed from widespread stratigraphic evidence, is represented by four of **its** six members within a single commercial deposit of Cheltenham refractory clay in Missouri. Scan electron micrographs show progressive changes in texture from plastic refractory clay (as in ball-clay "swirl" texture) through semi-plastic, semi-flint, to typical flint clay (recrystallized, well-ordered kaolinite). Micrographic evidence supports the interpretation of the origin of the Cheltenham clay earlier made from field and macroscopic evidence. Source material from nearby residual, weathered clay was transported into paludal basins, "digested," partly recrystallized to kaolinite, brecciated and reconsolidated, essentially completed before being covered by younger Pennsylvanian-age sediments,

Key Words: Cheltenham, Flint-clay, Kaolinite, Refractory.

INTRODUCTION

A flint-clay facies was originally proposed from stratigraphic, geologic field evidence observed across a regional span of about 150 km. In this paper an example will be illustrated, with scan electron micrographs (SEM's), where most of the members of the flint-clay facies occur within a single pit (less than 100 m wide) opened in the Cheltenham clay in central Missouri. In other words, a small example illustrates internally that which is also shown over a much larger, external occurrence—as if an inorganic "ontogeny recapitulates phylogeny."

The initial example of a flint-clay facies also was drawn from the Pennsylvanian-age Cheltenham formation which is widespread on the northeastern flank of the Ozark Dome in Missouri. The facies was named from the flint-clay member which is "characteristically a facies member in a non-marine mudstone sequence that includes high-alumina minerals, or potentially so, grading through flint clay into plastic clay, and extending into marine shale" (Keller, 1968).

Since the time when the flint-clay facies was proposed, several pits have been opened in which major representation of the facies members occurs within those individual pits. In the pit to be described commercial quantities of the sequence, plastic refractory clay, semi-plastic, semi-flint, clear flint clay, accessory "coaly soft-flint," and brecciated flint clay have been mined. End members of the regional example, not present in this pit include high-alumina minerals up-dip, and marine shale down-dip (Keller, Westcott and Bledsoe, 1954).

THE DEPOSIT AND SAMPLING

Clay samples were collected (courtesy of Alsey Refractories Co.) from the Alsey Pit located on Missouri Highway 161 about 4 km north and east of Middletown (near the north border of Montgomery Co., Mo.). The deposit in which this pit is opened is representative of the karstic, paludal sedimentary basins in which the Cheltenham refractory clay occurs (Allen, 1937; Bradley and Miller, 1941; McQueen, 1943; Keller, Westcott and Bledsoe, 1954; Keller, 1968).

Briefly; the Cheltenham "fire clay" in this pit, which is typical, grades downward through a sometimes pyritic, sandy, chert breccia, long referred to the Graydon formation (McQueen, 1943), but more recently reviewed in stratigraphical nomenclature less specifically by Howe and Koenig (1961). This chert is residual from the weathered portion of the underlying, cherty, Mississippian Burlington limestone. The upper surface of the weathered Burlington is undulating, such as characterizes a solution-corroded, somewhat karstic surface of weathering. Because of the irregular floor, the thickness of the Cheltenham clay varies across the pit (which was uncovered for about an acre in area). A working face of clay some 3 m to 4 m in height was sampled. The commercial-quality refractory clay is overlain by greenish (iron-rich), plastic, low-fusion, nonrefractory clay and shale which, in turn, is overlain by Kansan glacial drift.

A corelike body of clear flint clay (now mined) merged gradationally within a few centimeters into semi-flint clay which then graded into semi-plastic and plastic refractory clay. In several so-called "plastic" pits near Fulton and Mexico, Mo., where flint clay is also present as a minor constituent, the flint clay occupies the basal part of the deposit, and grades downward into the sandy phase of the Graydon member. In general in the north Missouri district the more flintlike clay is in the lower part of the deposit and grades upward into the more plastic clay toward the top. As a general rule, the semi-flint, semi-plastic, and plastic clays do not occur as uniform, sharply marked, or welldefined layers or zones. Instead these lithologies grade irregularly one into the other, so that selective mining must be strictly practiced in order to maintain quality control of the clay product. My interpretation of the

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Fig. l. Samples of refractory clay representative of members of the flint-clay facies. From left to right, they are plastic, semi-plastic, semiflint, flint clay, and brecciated flint clay. All samples are from a single deposit, the pit of the Alsey Refractories Co.. near Middletown, Mo.

Fig. 2. SEM of plastic refractory clay, $5000 \times$. The anhedral kaolin flakes in face-to-face orientation possibly represent floccules of clay sediment contributed by an earlier erosional cycle. Certainly they are not crystal stacks of kaolinite such as might weather *directly* from feldspar. The length of the bar at the figure number represents $1 \mu m$ unless otherwise designated by a number. All of the clay specimens micrographed were broken from the respective lumps shown in Figure 1.

Fig. 3. SEM at 3000× of the same specimen as Figure 2. Two orientations of clay flakes are shown within one field of view: a central kernel with flakes normal to the photo surface, and nearly flat-lying flakes in the lower right. This differs from the texture of fissile shales in which the clay flakes tend to be more uniformly oriented.

Fig. 4. This SEM shows the gross swirl pattern typical of ball clay, 1000×. Same hand specimen as in Figures 2 and 3.

Fig. 5. X-ray powder diffractograms: A, plastic clay; B, flint clay. Degrees 20 along the base. Scan rate, 2° per min; supplementary trace at left, $\frac{1}{4}$ per min between 21° and 22°. Cu-radiation.

geologic genesis of this consistent local variability, within the generality of harder clay below becoming softer, more plastic upward, includes several interacting processes which will be reviewed after the SEM's are examined.

Clay samples for micrography were collected from parts of the mine face in which lithologies typical of the facies members were developed (Figure 1). The lithologic differences in these refractory clays may not be easily discernible to a first observer but, as is known from other examples of selective mining, experienced miners can grade "fire clay" with astonishing accuracy by using subtle textural and surficial appearances quite evident to their trained eyes.

Clay samples were X-rayed (powder diffraction method) not only in pulverized form, but in smoothed slices cut from clay lumps. The inherently tiny crystals of clay minerals in these refractory clays, oriented randomly in the clay rock, make such slices truly representative of the clay in its natural occurrence for goniometer diffractometry. For SEM, freshly broken clay surfaces (no artifacts) were lightly coated with gold and micrographed.

SCAN ELECTRON MICROGRAPHS

Scan electron micrographs of the plastic clay are shown in Figures 2–4, at $5000 \times$, $3000 \times$, and $1000 \times$, respectively. The platy, anhedral, bent and twisted flakes of this plastic clay produce the "swirl" pattern that is a texturally identifying characteristic of ball clay (the ultraplastic kaolin clay)-reported and illustrated in earlier papers (Keller, *1976, 1977,* 1978 in press). It is interpreted, on a basis of SEM texture, that this plastic refractory clay was a ball clay in Pennsylvanian time; it has lost some plasticity and possibly been increased in refractoriness by diagenetic properties since the Paleozoic.

Its X-ray powder diffractogram, Figure 5A, shows it to be a poorly ordered kaolinite with a small amount of accessory quartz (common in the plastic varieties of the Cheltenham clay). The main diffractogram, run at 2° per min, is supplemented, on the left, by a short trace between 21° and 22° 2 θ run at 1/4° per min to show detail in the region of 4.18 and 4.13 \AA spacings. Bridley (1961) has suggested, "the partial resolution of the double $11\overline{1}$ and 111, with spacings 4.18 and 4.13 \AA as an indication of a well crystallized kaolinite In diffractometer records the 4.13 \AA peak gives a step on inflection on the side of the 4.18 Å." Obviously, neither two peaks nor inflections are resolved—the plastic clay is poorly ordered.

Semi-plastic (less plastic) clay is micrographed in Figures 6 and 7, at $3000 \times$ and $1000 \times$, respectively. Note that the "swirl" pattern present in the plastic clay

Fig. 6. SEM of semi-plastic refractory clay, 3000×. Compared to the plastic clay, this semi-plastic is coalescing to tighter, more nearly equidimensional aggregates.

Fig. 7. SEM, at 1000×, from same hand specimen as Figure 6. The swirl pattern is more subdued and giving way to newly developing crystal aggregates.

Fig. 8. SEM of semi-flint clay, 3000×. Almost all of the original swirl pattern indicative of many thin original plates is lost as kaolinite re-formed into aggregates of less-plastic clay.

Fig. 9. SEM at 1000× of same specimen as Figure 8. Note in lower left third of photo apparently newly developing (recrystallization) kaolinite crystals.

Fig. 10. SEM of flint clay, 5000×. A very compact mass of interlocking, neo-formed and/or recrystallized kaolinite. The packets are thin stacks of kaolinite plates comprising new crystals of kaolinite.

Fig. 11. The left center of Figure 10 doubled in magnification to 10,000x shows detail of new kaolinite plates making up the interlocking packets.

is losing curvature and giving way to "flatter," incipient stack-piled platy crystals such as will be more completely developed in flint clay. The X-ray diffractogram is not significantly different from that of the plastic clay: it has not been reproduced.

Semi-flint clay is micrographed at $3000 \times$ and $1000 \times$ in Figures 8 and 9, respectively. Almost all of the swirl pattern has been lost. The clay plates are stacked flatter than in the more plastic clays.

A textural pattern giving a hint of developing kaolin crystals, analogous to the appearance of incipiently developing crystals at low metamorphic grade in knotenschiefer (spotted slates), may be seen in the lower center of the micrograph. Presumably diagenesis has been progressively more effective and prominent in the semiflint clay than in the two previously discussed.

Clear (homogeneous, commercial-quality) flint clay is micrographed at $5000 \times$ and $10,000 \times$ in Figures 10 and 11, respectively. In Figure 10, the packets of tiny kaolinite plates are well developed, interlocking, and tightly intergrown with one another in random orientation. These observations are more convincingly substantiated at the higher magnification in Figure 11.

The X-ray diffractogram of this flint clay, Figure 5B, shows a notably higher ordered kaolinite than any of the more plastic clays. A slight shoulder is present on the $21^{\circ} - 22^{\circ}$ trace, which also is notably higher in intensity than counterparts of the preceding clay members. No quartz is present in this sample of flint clay. Quartz commonly occurs near the contact of flint clay with the pit rims, or sporadically within some deposits, but "sandy" flint clay is not mined for refractory purposes.

These SEM's of flint clay aid in explaining the distinguishing *lithologic* characteristics of it (flint clay) in hand specimen: namely, its remarkable homogeneity, fine-grain or crystal, and conchoidal fracture (a manifestation of homogeneity of fine texture). These macroproperties are due to the micro-, internal makeup of the clay: a myriad of relatively uniform, randomly oriented, usually well-ordered crystals of kaolinite which was newly crystallized and recrystallized from the "digested" sediment in the old swampy basin. Flint clay is a monomineralic rock which exhibits nonclastic crystallinity, therefore, texturally it resembles a microsyenite more closely than a shale.

The texture of flint clay obviously is strikingly different from that of plastic clay, Figures 2-4, although both are from the same deposit. They were collected only a few meters apart, with no visible sharp break in stratigraphy, process, or time, as otherwise might have indicated a change in either provenance or sedimentational regime. From field observations and ceramic properties (ceramic compatibility between flint clay and associated plastic clay) it has been concluded that they have a conjugate origin (McQueen, 1943; Keller, Westcott and Bledsoe, 1954; Keller, 1968).

The brecciated flint clay, Figure 1, likewise is wellordered kaolinite. The texture of the pelletal clay is only slightly finer grained than the matrix, a difference scarcely detectible at $5000 \times$, so it is not illustrated, nor is particular significance attached to it. The breccia indicates, however, in my interpretation, that the major clay-forming processes had been completed by the end of Cheltenham time. Flint clay had been formed, brecciated and again lithified. This relationship accords with the geologic evidence from the Bucker diaspore pit 100 km south. There the refractory clays had been formed and partly weathered (a terrestrial process) in Pennsylvanian time before the incursion of marine conditions which followed. Marine rocks overlying the weathered flint clay are, in sharp contract, the fresh, green illitic Excello (Pennsylvanian) shale, and the thick, marine Fort Scott limestone (Keller, 1952, 1968). The dominant clay-forming processes were completed before being covered by the marine sediments. The source sediment for the refractory clay in the Alsey pit most probably was illitic and kaolinitic soil-clay residual from the weathering of the surrounding country rock at that time, the Burlington limestone (Robbins and Keller, 1952). After and/or during deposition further kaolinization, desilication, and potassium removal from illite in the low-lying Pennsylvanian-age swamps was interpreted to have taken place by upward and lateral leaching in a mud-gel state via the Donnan effect (Keller, 1968). If leaching had been primarily downward, as in highland topography, the "parent" kaolinite (flint clay) would have been in the uppermost (freshest water) zone. Instead, the flintier, higher-fusion clay is found in the lower zones of the Cheltenham clay deposits in Missouri.

After younger Pennsylvanian sediments covered the Cheltenham clay, dewatering and settling and compaction occurred and commonly generated movement that produced slickensides in the clay and irregular and gradational zonation within the clay body (deposit). Continuing diagenetic changes operated to further consolidate and lithify the clay.

CONCLUSIONS

Scan electron micrographs yield graphic laboratory evidence of (1) a flint-clay facies developed in a single, Cheltenham refractory clay deposit in Missouri, and (2) the graphic evidence supports earlier-garnered, geologic evidence from field evidence for the origin of that clay.

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REFERENCES

- Allen, V. T. (1937) The Cheltenham clay of Missouri: *Mo. Geol. Surv. 59th Bienn. Rep.* Appendix V, 29 pp.
- Bradley, R. S. and Miller, B. K. (1941) Prospecting, developing, and mining semi-plastic fire clay in Missouri: AIME *Tech. Pabl.,* No. 1328, 9 pp.
- Brindley, G. W. (1961) *Kaolin, Serpentine, and Kindred Minerals in the X-ray Identification and Crystal Structures of Clay Minerals* (Edited by Brown, G.): Mineral. Soc., London, pp. 51-131.
- Howe, W. B. and Koenig, J. W. (1961) The stratigraphic succession in Missouri: *Mo. Geol. Surv. Water Resour.* 40, 2nd ser., 185 pp.
- Keller, W. D. (1952) Observations on the origin of Missouri high-alumina clays: AIME Symp. Vol., Problems of Clay and Laterite Gen*esis,* pp. 115-135.
- Keller, W. D. (1968) Flint clay and a flint-clay facies: *Clays & Clay Minerals* 16, 113-128.
- Keller, W. D. (1976) Scan electron micrographs of kaolins collected from diverse environments of origin---II: *Clays & Clay Minerals* 24, 114-117.
- Keller, W. D. (1977) Textures of kaolin-rich refractory clays as shown by scan electron micrography: AIME preprint No. 77-H-302, St. Louis, Oct. 1977, 17 pp.
- Keller, W. D. (1978) Classification of kaolins exemplified by their textures in scan electron micrographs: *Clays & Clay Minerals,* 26, $1 - 20$.
- Keller, W. D., Westcott, J. F. and Bledsoe, A. O. (1954) The origin of Missouri fire clays: *Clays & Clay Minerals, Proc. 2nd Conf.* N.A.S.N.R.C. Publ. 327, 7-46.
- McQueen, H. S. (1943) Fire clay districts of east Central Missouri: *Mo. Geol. Surv. Water Resour.* 2nd Ser., 24, 250 pp.
- Robbins, C. and Keller, W. D. (1952) Clay and other noncarbonate minerals in some limestones: *J. Sediment. Petrol.* 22, 146-152.

Pe3юмe- Фации твердых огнеупорных глин, первоначально предполагаемые по региональным стратиграфическим данным, представлены 4 из 6 их типов в одном и TOM же промышленном месторождении Челтенхамской огнеупорной глины в штате Mиссури. Снимки, сделанные электронным развертывающим микроскопом, показывают прогрессивные изменения в структуре глины: от пластичной огнеупорной глины /как в "водоворотной" структуре комовой глины/ к полу-пластичной, полу-твердой глине и затем к типичной твердой огнеупорной глине/перекристализованный, xopomo упорядоченный каолинит/. Микрографические данные подтверждают предположение о происхождении Челтенхамской глины, высказанные ранее по резуль-TaTaM полевых и макроскопических исследований. Исходный материал из окрестной остаточной, выветренной глины был перенесен в болотистые впадины, преобразован, частью перекристаллизован в каолинит, брекчирован и вновь консолидирован, т.е. существенно сформирован до того, как был перекрыт более молодыми осадками Пенсильванского возраста.

Kurzreferat- Flint-Ton Fazies, die ursprünglich wegen weitverbreiteter, stratigraphischer Beweise vorgeschlagen wurde, ist durch 4 ihrer 6 Mitglie der innerhalb einer einzigen, gewerblichen Ablagerung von Cheltenham, feuerfestem Ton in Missouri, vertreten. SEM zeigt fortgeschrittene Veränderun gen in Struktur-von plastischem, feuerfestem Ton (wie in plastisch-Ton "Strudel" Gefüge) bis zu semi-plastisch, semi-Flint und zu typischem Flint-Ton (umkristallisiertes, übersichtliches Kaolinit). Mikroskopische Daten unterstützen die Interpretation von der Abstammung des Cheltenham Tons, welche schon früher durch makroskopische und Feldbeweise gemacht wurde. Ursprüngliches Material von nahem, übriggebliebenem, verwettertem Ton, wurde in moorige Becken gebracht,"verdaut", zum Teil umkristallisiert zu Kaolinit, Brekzien geformt und wiederkonsolidiert, im Großen und Ganzen vervoilstandigt bevor es mit Pennsylvania-altem Sediment bedeckt wurde.

Résumé-Le facies "flint-clay", proposé à l'origine à partir d'évidence stratigraphique étendue, est représenté par 4 de ses 6 membres d'un seul dépôt d'argile réfractaire de Cheltenham du Missouri. Les micrographies électroniques montrent des changements progressifs en texture d'argile réfractaire plastique(comme dans la texture "tournoyante" d'argile-boule), à la texture demi-plastique demi-flint, et finalement à la texture typique de flint-clay (une kaolinite recristalisée,bien ordonnée).L'évidence micrographique appuie l'interprétation de l'origine de l'argile de Cheltenham déduite plus tôt de l'évidence obtenue sur le champ et de l'évidence macroscopique. Du matériel de source d'un argile proche, résiduel et altéré, a été transporté dans des basins paludéens,"digéré",partiellement recristallisé en kaolinite,brisée et reconsolidée, essentiellement complétée, avant d'être recouverte par sédiments plus jeunes d'âge Pennsylvanien.