GEOLOGY AND MINERALOGY OF THE SEDIMENTARY KAOLINS OF THE SOUTHEASTERN UNITED STATES—A REVIEW*

by

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ABSTRACT

Beginning with the work of Ladd in 1898, much has been written about the kaolins of the Southeast, with maximum coverage on the deposits of greatest economic value. Early studies were, of necessity, general in nature but also contain excellent detailed descriptions. In the past 15 years increased demands for kaolins to meet specific requirements have resulted in a large amount of detailed research, much of it unpublished. This paper attempts to review the available information placing emphasis upon the geologic and mineralogical data and the interpretation thereof with respect to the occurrence, detailed character and variability, and probable origin of the clay deposits.

The commercial deposits of kaolin clay discussed herein lie throughout the sands of the Upper Cretaceous Tuscaloosa formation and are localized in a narrow belt along the southeast edge of the "fall line" from Macon, Georgia, to Aiken, South Carolina. The kaolin lenses are irregular in shape, size and purity, ranging from a few feet to a mile in length, and up to 50 ft in thickness. Variability within the lenses is a function of (1) the concentration and localization of non-clay materials (quartz, mica, gibbsite, pyrite, lignite; Fe_2O_3, TiO_2, MnO), (2) the montmorillonite content, (3) the particle size and X-ray crystallinity of the kaolinite, and (4) the texture of the clay aggregate. These mineralogical, chemical and textural variables are not homogeneously distributed but reveal and help to explain differences between samples, layers, pits and the important hard and soft clay types of the region.

INTRODUCTION

As pointed out by Heinrich Ries (1927) in his famous work *Clays, their occurrence, properties and uses*, "The term kaolin was originally used to refer to white residual clays of a white or nearly white burning character, but in recent years it has been stretched to cover certain white sedimentary clays like those obtained in South Carolina and Georgia." I have used the term kaolin, rather than kaolinite, in the title of this review not only in recognition of former practice, but also because it points to the fact that

* Contribution No. 63–60 from the College of Mineral Industries, The Pennsylvania State University.

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the clays to be discussed are not monomineralic. Much of the variability between and within the deposits is the result of variation in the nature and quantity of minerals other than kaolinite, such as mica, quartz, varieties of TiO_2, pyrite and the clay mineral montmorillonite. The common practice of beneficiating the clays by washing is testimony to their naturally impure state even though, as so ably expressed by Veatch (1909, p.101), "it would seem that nature operated a clay washing plant on a grand scale" in the original separation of the fine from the coarse fraction of the enclosing sands.

The clay mineral kaolinite, though not infrequently found as pseudo-morphs after a wide variety of silicate and even non-silicate minerals, is, on a quantitative basis, believed to be derived largely from the weathering of feldspar and Al-mica. It occurs in all types of sedimentary rocks and is particularly common in association with arkoses and the related silts and shales (Krynine, 1948, p.149). A sedimentary deposit of commercial importance may be produced where conditions are favorable for an adequate amount of the clay to be separated from associated non-clay material, deposited without appreciable "dilution" by other detritus or chemical precipitates and preserved from subsequent alteration or erosion. It is to be expected that for every "commercial deposit" there will be many associated non-commercial clay concentrations having all degrees of purity and ranging from miles to feet in size.

Since the literature on the sedimentary kaolins of the southeastern United States is concerned primarily with the commercial clays, any review of the literature presents only part of the entire geological and mineralogical picture. This particular review is further limited to a discussion of the kaolins in the Upper Cretaceous Tuscaloosa formation of Georgia and South Carolina.

LITERATURE

Although reference to the clays under consideration is to be found in early geological studies of the region, the first detailed description was that of George E. Ladd published in 1898 as a Georgia Geological Survey bulletin. In Georgia his work was followed by the comprehensive studies of J. O. Veatch (1908, 1909), while in South Carolina a preliminary report was made by Sloan (1904). The work of Veatch stands, even today, as an authoritative, informative description of the geology and mineralogy of the exposures and pits accessible in the early days of the century; and many of his concepts as to the origin of the clays have been subscribed to by later workers. While Veatch was occupied with the clays, his colleague L. W. Stephenson (1911, 1914) was engaged with the problem of placing them in proper stratigraphic relationship to associated lithologic units, a task undertaken in earlier days by Smith and Johnson (1887) who had originally named the Tuscaloosa formation.

Ries (1903, 1922, 1927) described, in increasing detail, the Cretaceous coastal plain clays of Georgia and South Carolina, and put them in proper perspective with other clays in these and other states with respect to their known geographic position, geologic occurrence, apparent mineralogical character and commercial importance.

With the development of the kaolin industry and greater recognition of its economic importance to the region, came an increasing amount of work, which has continued to the present day, on the exploration, properties, processing, and utilization of the clays (e.g., Weigel, 1922, 1925; Stull and Bole, 1936; Henry and Vaughan, 1937; Lang, 1940; Thompson, 1943; Warren and Thompson, 1943; Klinefelter *et al.*, 1943; Kesler, 1951; Woodward, 1955; Murray, 1963). A recently issued bibliography on "High alumina kaolinitic clay of the United States" (Mark, 1963) contains many useful references pertinent to the clays considered here.

Concurrent studies emphasizing stratigraphic relationships but often including descriptions and discussions of probable origin of the clays have been numerous. Among them are the following: Cooke, 1926, 1936, 1943; Neumann, 1927; Smith, 1929; Stephenson *et al.*, 1942; Munyan, 1943; LaMoreaux, 1946; Eargle, 1955; and Le Grand and Furcron, 1956.

Discussions of the mineralogy of the clays and associated sediments and chemical analyses of the clay are to be found in many of the references already given, as for example in Veatch (1908, 1909), Stull and Bole (1926), Neumann (1927) and Smith (1929). The classic paper of Ross and Kerr (1930) revealed the applicability of the X-ray diffraction technique to the study of the kaolin minerals and discussed many aspects of their characterization and formation. Although only one sample from the Georgia–South Carolina belt was included, this work provided much information basic to subsequent studies on these kaolins. Particularly noteworthy, among such studies, have been those of Klinefelter, O'Meara, Truesdell, and Gottlieb (1943), Mitchell and Henry (1943), Kerr and his associates on Research Project 49 of the American Petroleum Institute (1949, 1950), and Hinckley (1961, 1963).

The Bureau of Mines publication by Klinefelter *et al.* (1943) reports the results of the comprehensive analysis of 18 selected specimens by mineralogical, chemical and physical testing procedures. This work provided important evidence as to differences between the hard and soft clay types of the region, as represented by a limited number of samples. Independent work of Mitchell and Henry published in the same year (1943) also revealed differences between hard and soft samples, and, together with the previous work cited, may be considered a major step forward in the continuing attempt to relate physical behavior of the clays to their mineralogical and chemical properties.

The work of contributors to American Petroleum Institute Project number 49 resulted in a mass of mineralogical and chemical data on a large number of clay specimens, including four samples from the Macon,

Georgia, area and four from Bath, South Carolina. The undertaking was not only the first attempt to characterize each of the samples by a combination of all modern mineralogical tools, but also provided a thorough survey of work done previously by others in each area, and reviewed proposed theories of origin. The various reports are referenced under Kerr and Hamilton (1949); Kerr and Kulp (1949); Kerr, Kulp and Hamilton (1949); Holmes (1950); Main, Kerr and Hamilton (1950); Davis *et al.* (1950); Kerr *et al.* (1950); and Adler *et al.* (1950).

Variations in the X-ray crystallinity of the Georgia clays were measured by Klinefelter *et al.* (1943) and studied in detail by Murray and Lyons (1956, 1960) and Hinckley (1961, 1963) in work relating crystal perfection to various other properties.

The most comprehensive mineralogical study of the kaolins, involving the measurement of 20 variables and 343 samples, was completed by Hinckley (1961; see also Hinckley and Bates, 1960, and Hinckley, 1963). This work was done on material from nineteen drill cores taken in nine pits and supplied by several of the kaolin companies in the area. The more important results will be reviewed in succeeding pages.

Finally, for a thorough evaluation of both the geological and mineralogical characteristics of the deposits as studied prior to 1956, plus a new approach to various aspects of the theories of origin, the reader is referred to the article of Kesler (1956) which was reissued with revisions for the benefit of those attending the conference covered by the present volume (Kesler, 1963).

GEOGRAPHIC AND GEOLOGIC SETTING

The sediments of the Atlantic Coastal Plain contain vast amounts of clay much of which is kaolinite. In areas where natural sorting processes operated most efficiently, kaolin deposits of high purity have resulted. Those of greatest commercial importance are the result of an appropriate combination of many factors, the most significant being the size and homogeneity of the clay lens, the clay mineral composition and purity, and, from the mining point of view, the nearness of the lens to the present surface (amount of overburden). The known deposits which best meet these conditions have been found between Macon, Georgia, on the south and Aiken, South Carolina, to the north lie in the Upper Cretaceous sands which outcrop in a narrow belt along the southeast edge of the "fall line". Other important deposits occur outside of these geographic and geologic limits but will not be dealt with in this paper.

In the area being discussed herein, the Upper Cretaceous Tuscaloosa formation, which contains the clay lenses under consideration, rests unconformably upon underlying crystalline rocks and, except where exposed near the "fall line" by erosion, is overlain by the Upper Eocene Barnwell

formation. The nature of these geological units and their stratigraphic relationships with other formations has been thoroughly discussed by workers already referenced (in, for example, Kesler, 1956, or La Moreaux 1946).

The underlying crystalline rocks—as revealed (1) in drill cores, (2) in the present Piedmont and (3) where overlying Coastal Plain sediments have been removed by erosion—consist of a wide variety of igneous and metamorphic rocks assigned to ages from Pre-Cambrian through the Paleozoic and intruded, in part, by Triassic dikes of diabase. In discussing the regional geology of the Blue Ridge and Piedmont provinces, Espenshade and Potter (1960, p.5) point out that: "Two general types of metamorphic rocks predominate in the region: hornblende schists and gneisses and micaceous siliceous schists and gneisses. . . . Large granite bodies occur in parts of the region and pegmatites are abundant in certain areas. Gabbro and diorite are common; small bodies of ultrabasic rocks occur mostly in the Blue Ridge area. A large area in central North Carolina is underlain by volcanic flows, tuffs, and slates. . . . These rocks extend southwest across South Carolina into Georgia. . . . Isolated basins of unmetamorphosed sedimentary rocks of Triassic age occur in the Piedmont of Virginia and North Carolina."

The ability of Blue Ridge and Piedmont rocks, of the type now exposed there, to serve as the source of large quantities of kaolin minerals has been well established by detailed evaluation in specific areas (e.g., Cady, 1950, and Sand, 1956), and by more recent studies of the soils throughout much of the region. Particularly significant in this latter respect is work of the type sponsored by fourteen agricultural experiment stations and the U.S. Department of Agriculture as part of a cooperative regional research program (Anonymous, 1959). In 22 profiles of Piedmont soils formed on both acid and basic rock types from Georgia through Virginia, kaolinite (including varying amounts of halloysite) was the most abundant clay mineral in the C horizon of all but one profile and persisted as the most abundant clay throughout the entire profile in sixteen of the locations studied. Vermiculite was found to be the next most abundant clay mineral and montmorillonite was present as a minor component in seven of the twenty-two profiles.

Lying unconformably on the Tuscaloosa formation are the Barnwell sediments of Upper Eocene age. The following description is quoted from Kesler (1956, p.546):

> "Mining and exploratory drilling have shown that the Barnwell in central Georgia consists of two fairly uniform units. The lower is the Twiggs clay member, which is 80 to 115 feet thick and consists of Fuller's earth containing limestone beds that are most prominent near the base, as shown in Fig. 4. The lowermost bed is very sandy and commonly about four feet thick, and is leached to loose sand along the outcrops. These limestone beds are highly fossiliferous, and represent an interfingering of the Ocala limestone from the southwest. A dorsal vertebra of the "zeuglodon" *Basilosaurus cetoides*, identified by Remington Kellogg, was found by the writer

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in 1951 in these beds at Rich Hill in Crawford County, 20 miles southwest of Macon.
The upper member of the Barnwell is the Irwinton sand, an almost pure sand at least 50 feet thick containing a few random thin beds of Fuller's earth. Where least weathered, the sand is weakly indurated. LaMoreaux described a residual deposit of coarse red sand 20 feet thick overlying the Irwinton sand, and regarded it as possibly a third member of the Barnwell. It appears, however, that this deposit is a part of the colluvial mantle of later origin . . .''

Brindley (1957) has shown that fuller's earth obtained in the Twiggs clay member of the Barnwell in the vicinity of Dry Branch, Georgia, is "a mixture of montmorillonite and cristobalite in comparable proportions, with minor amounts of quartz and traces of mica and kaolinite".

The Tuscaloosa formation, in the area of concern, strikes northeast–southwest and dips southeastward about 15 ft per mile (LaMoreaux, 1946, p.44), thickening down-dip. As described by Veatch (1909, p.93):

"The Tuscaloosa formation consists entirely of sands, gravel and clays, generally unconsolidated. . . . The sand . . . is composed principally of quartz in small angular particles; muscovite mica is next the most abundant mineral, and small amounts of hornblende or augite, garnet and magnetite have been observed. Feldspar in various stages of decomposition occurs, while the sands are often colored by iron oxide, limonite, and hematite. The sand is generally coarse grained, and near the contact with the crystalline rocks, beds of gravel, and large subangular fragments of quartz occur."

Neumann (1927, p.378) adds the following heavy mineral constituents to those mentioned above: andalusite, biotite, cassiterite, gold, kyanite, ilmenite, leucoxene, magnetite, monazite, rutile, sillimanite, titanite, tourmaline and zircon.

The sands are commonly cross-bedded and contain many minor disconformities. These characteristics plus the angular character of the quartz, presence of feldspar grains equal in size to the quartz, and scarcity of rounded gravel led Kesler (1956, p.545) to the belief that the "series reflects rapid deposition without sorting of the clastic material according to size or density".

Kaolin is found in the sand in amounts ranging from pseudomorphs after single feldspar crystals to the huge lenses of commercial importance. Contacts between clay concentrations and enclosing sands vary from sharp to gradational, the latter being predominant. Furthermore, the argillaceous character of much of the sand itself is made apparent by sections and descriptions recorded throughout the literature as illustrated by the following sections from Main, Kerr and Hamilton (1950, pp.20 and 21):

	Birch Pit Macon, Ga.		Dixie Rubber Pit Bath, South Carolina
2–4 ft	Sandy soil	10–15 ft	Red sand and soil
10 ft	Red clay and soil	10–30 ft	Sand cemented with about 10 per cent kaolin
2–10 ft	Fuller's earth		
8–10 ft	Sand. Contains marine fossils	50 ft	White angular sand, Clay at bottom
10 ft	Ocala limestone—contains marine fossils	10 ft	High grade kaolinite

10–15 ft	Kaolinite	5 ft	Kaolinite with quartz
12–20 ft	Sand with clay laminations and clay cement	?	Sandy, clay-like sediment
15 ft	Gray to brown clay	6–10 ft	Gray sand, leached
10–15 ft	Kaolinite	15–20 ft	Sand, cemented with clay
?	Sandy clay	30 ft	Sand, red, with clay at bottom
		10–20 ft	Kaolinite

Such sections indicate that nature's washing plant operated with varying efficiency producing on occasion nearly perfect separation of coarse and fines and thus "clean" sands or "pure" kaolins, but more often yielding sandy clays or clayey sands. Since major interest in the Tuscaloosa has naturally centered about the "best" clays and the sands adjacent thereto, it is unlikely that the literature gives us a representative picture of the formation as a whole.

THE KAOLIN DEPOSITS

Associated Features

The clay lenses in the Tuscaloosa range up to about 50 ft in thickness and more than a mile in length. Irregular in outline, they tend to be elongate in plan and lens-shaped in cross-section.   Well logs indicate that they occur at various depths throughout the Tuscaloosa.

Associated features visible in the outcrop or the exposed faces of the pits include thin lenses and seams of lignite, nodules of pyrite occurring separately or in vertically contiguous groups, interpenetrating layers of sand, and—where exposed to surface weathering—yellow-brown streaks and layers due to oxidation of iron. Bauxite has been found associated with the clays in three types of occurrence: (1) as boulders in the soil above kaolin lenses stripped by erosion of their Tertiary cover; (2) in beds at or near the top of lenses truncated either by the present surface or the Cretaceous–Tertiary unconformity; and (3) "as sporadic bodies . . . found, in drilling, as much as 30 ft below the unconformity and invariably in the interior of kaolin deposits . . . containing no clear evidence of subaerial erosion . . . " (Kesler, 1951). Types 1 and 2 are thoroughly described by Veatch (1909, pp.430–447) and type 2 is of particular interest because of the common association with an indurated, porous, and sometimes nodular variety of kaolin clay locally known as chimney rock because of its former use in building chimneys. Where observed by the writer (e.g., seven miles north of McIntyre at the Joe Boone pit of the Chattahoochee Brick Company) this clay lies between the bauxite and the present soil cover and varies considerably in thickness due to a sharply undulating contact with the bauxite.   Although very hard where exposed on the

stripped surface at the edge of the pit and in loose blocks broken out in former mining operations, the material in a freshly exposed face becomes more soft and plastic as the bauxite contact is approached.

Kesler (1956, pp.550–551) suggests that sulphuric acid provided by the action of meteoric waters and pyrite-rich lenses in the clay may have played an important role in the bauxitization process, thus explaining not only the bauxite lenses found near an erosion surface but also those occurring in the interior of uniform, thick kaolin lenses well below and apparently unaffected by an unconformity. The reader is referred to Kesler's paper for suggested details of the alteration process.

Variability within the Clay

Physical characteristics.—The obvious variation in clay samples taken from different pits and different localities within the region is in their so-called hardness in the air-dried condition. Stull and Bole (1926) first used the terms hard, semi-hard, and soft as part of their classification scheme for the Georgia kaolins; and although, with the development of the many and varied modern uses of the clay, this classification has lost some of its original significance, the distinction between hard and soft types appears to be significant in the light of other physical, chemical and mineralogical properties and considerations as to origin. The justification of an inter-mediate or "semi-hard" category is debatable in the light of conclusions by both Klinefelter *et al.* (1943) and Hinckley (1961) that, on the basis of the samples studied, other properties suggest the existence of separate hard and soft groups rather than a single population with hard and soft end-members. Field studies support this contention in that pits are usually described in the literature as containing either hard or soft clay rather than a mixture, although Kesler (1956, p.549) reports that "some deposits consist of two beds, one 'soft' and the other 'hard' with a plane of contact between them". Although it has been claimed in the past that hard versus soft clay types show preferred lateral and/or vertical distribution through-out the region, extensive drilling and development in the area suggests that such is not the case but that definite conclusions cannot be drawn without a complete compilation and study of all available data.

An association of hard and soft clay types with particle size differences has been noted by a number of the investigators. Even though different workers have used a variety of measurement techniques and often have quite different concepts as to the definition of a "particle", it seems well established that hard clays have smaller particles than soft. Mitchell and Henry (1943), for example, obtained grain-size distribution curves on samples deflocculated under optimum conditions and found that 80 per cent of the particles in the hard clay but only 50 per cent of those in the soft were smaller than 2μ. Hinckley (1961, p.87), by electron microscope

measurements on eight samples from two hard clay pits and eight others from two soft clay deposits, showed that the association of small particle size with the hard type of clay is statistically significant. In this work measurements were made on replicas of fractured surfaces and recognition of separate "particles" was predicated on the tendency of the material to break around rather than across individual, morphologically contiguous crystallites.

As might be expected, textural variations in the kaolins are common. Most significant perhaps from the standpoint of genetic connotations are relationships observed in thin section. Thus Hinckley (1961) has shown that, whereas X-ray patterns of clay blocks with shaved, plane surfaces indicate random orientation of crystallites, observation of smaller areas seen in the microscope reveals that hard clays commonly have aggregate birefringence due to patches of clay particles in parallel orientation whereas the soft clays do not.

Related to the texture of the clay samples is the matter of the bulk density. Klinefelter *et al.* (1943) obtained an average value of 1.495 for the soft clay samples measured and 1.72 for the hard, with the so-called "semihard" samples giving results overlapping those from the hard and soft. Hinckley (1961, p.73) obtained a mean value of 1.48 for soft clays and 1.62 for hard. As this author points out, both these values are considerably less than the density of 2.60 for kaolinite.

Mineralogy.—Non-clay minerals occur in the kaolins as detrital grains, authigenic particles and minute inclusions in the clay particles.

The amount of detrital non-clay material in the clays is difficult to assess from the available literature because of (1) the variety of techniques used and (2) the fact that coarse fractions usually contain far greater amounts of undispersed clay aggregates and books than non-clay particles. The most accurate estimates are probably those of Main *et al.* (1950) obtained by microscopic examination and comparison with data from the chemical analysis. These indicate that even some of the purest kaolins contain up to 8–12 per cent of non-clay impurities of which quartz, mica, and feldspar are most important. Petrographic work on $>14\mu$ fractions by Klinefelter *et al.* (1943) revealed, in addition, the presence of rutile, zircon, tourmaline and occasionally calcite.

Minerals that are believed to have formed in the clays during and after their deposition are pyrite (commonly oxidized to limonite) and the titanium compounds usually referred to as leucoxene. Anatase is believed to be the most common constituent of the latter and $<1\mu$ particles of this mineral have been observed in electron micrographs of some of the clays.

Minute mineral inclusions are so characteristic of the typical kaolinite particles in these clays that their absence in the kaolin books is striking. Indeed, Klinefelter *et al.* (1943, p.12) describe the clay particles as either "clean" or "dirty", the latter being characterized by "minute opaques and

highly refracting particles (chiefly limonite, rutile, zircon, etc.)''. Although the nature, distribution, and concentration of these inclusions in the various clay types of the region have a very important bearing on the color and light scattering properties of the material, to the author's knowledge no detailed study of them has been published.

Of all the impurities present in the Georgia and South Carolina kaolins, probably the most important because of its bearing on the physical properties and therefore utilization of the clays is the clay-mineral montmorillonite. Its presence was not noted by early workers because of the small amounts present in most of the clays. Mielenz, King and Schieltz (in Kerr *et al.*, 1950, p.145) detected it by dye tests and confirmed it by X-ray diffraction. Murray and Lyons (1956, 1960) pointed out the importance of taking montmorillonite into account when considering the relationship of kaolinite crystallinity to paper-coating quality and other characteristics of the clays. Using a base exchange technique and X-ray fluorescence detection method Hinckley and Bates (1960a, 1960b) reported on the variation in montmorillonite content in 108 kaolin samples from six drill cores from three pits, two of soft-type clay and one of the hard-type. Values for individual samples ranged from 0.01 to 6.47 per cent in the soft clays and 1.00 to 3.16 per cent in the hard, but because of the great variability within the two clay types the difference in montmorillonite content between hard and soft clays was found to be not statistically significant.

Variation in the nature of the kaolinite itself has already been noted in the previous discussion of particle size and its apparent relationship to clay type. Related to the particle size is the degree of crystal perfection of the kaolinite as measured by X-ray diffraction. Klinefelter *et al.* (1943) first noted that certain X-ray diffraction peaks were not as well resolved in hard as in soft kaolin samples and pointed out (p.14) that ''the different conditions under which the hard and soft clays may have been formed could have caused concurrently the varying degree of crystalline development as well as some other conditions that have resulted in hardness or softness of the clays.'' In a later study in which 11 Tuscaloosa clays and two others were ranked on the basis of perfection of X-ray patterns and then compared from the standpoint of various other properties (Murray and Lyons, 1956), it is pointed out (pp.39, 40) that, in comparing the different samples, ''in general the coarser the particle size the better the crystal perfection'' whereas within any given sample, fine and coarse fractions show the same degree of crystal perfection. Hinckley (1963) reported on his measurement of the ''crystallinity index'' of 144 samples from eight cores and concluded that the four deposits represented ''are nonhomogeneous with respect to crystallinity and that the hard and soft types can be distinguished'' on this basis ''at the 0.95 probability level by an analysis of variance''.

The fact that the coarser-grained, better crystallized soft clays contain

a higher proportion of books than the finer-grained, more poorly crystallized hard clays has been noted by many observers. A statistically significant positive correlation between degree of crystallinity and abundance of books was found by Hinckley (1961, pp.105, 108). On the basis of the statements of Murray and Lyons, referred to above, as well as the microscopic and X-ray studies of others, it seems evident, however, that increased abundance of books is not, of itself, the cause of improvement of crystallinity values but is rather an indicator of conditions which resulted in better crystallinity in the sample as a whole. In considering the nature of such conditions and implications as to origin, it is also important to note that Hinckley (1961, p.38) recognized the existence of two types of books, namely those with a shaggy appearance believed to be formed from mica and others with smoother edges which make up a large majority of those occurring in the soft clays and are considered by most observers to be authigenic. Although as noted in a previous reference to the work of Klinefelter *et al.* the books represent the purest kaolinite in the deposits because of the absence of inclusions, it is only in recent years that the clay companies have taken any steps to reclaim them from the coarse fraction which, until then, had been totally rejected during the "beneficiation" process.

Chemical characteristics.—In addition to the major constituents SiO_2, Al_2O_3 and H_2O+, chemical analyses of typical, commercial kaolin samples before beneficiation usually show TiO_2 in excess of one per cent; MgO, CaO, Na_2O, and K_2O in the 0.01 to 1.0 per cent range; and $FeO+Fe_2O_3$ in the 0.10 to 2.0 per cent range. Variations between clay types are shown in the following data given by Hinckley (1961, p.57) for 50 hard and 197 soft clay samples measured by X-ray fluorescence. The differences between types are significant in the case of Al_2O_3 and Fe_2O_3.

		Mean %	Standard Deviation	Range
SiO_2	Hard	44.1	0.49	43.2–45.5
	Soft	43.8	0.75	41.1–47.9
Al_2O_3	Hard	38.6	1.07	35.4–40.7
	Soft	39.7	1.26	31.9–42.3
K_2O	Hard	0.28	0.19	0.02–0.76
	Soft	0.14	0.14	0.00–1.12
Fe_2O_3	Hard	1.9	0.71	0.75–5.52
	Soft	0.2	0.23	0.00–1.64
TiO_2	Hard	1.6	0.27	1.09–2.12
	Soft	1.5	0.48	0.43–3.87

Various trace element constituents have been measured by several workers. Klinefelter *et al.* (1943, p.7) note that "with a single exception boron is present in all of the soft clays. It is absent in eight of the eleven hard clays." Hinckley (1961, p.74), on the other hand, found more boron in eight hard samples than in a similar number of soft clays but showed that the difference was not significant due possibly in part to the small number of samples measured. Wheeler and Burkhardt (in Kerr *et al.*, 1950, p.85) point out that "the presence of zirconium and titanium in sizeable amount" in their samples from Georgia "indicates the sedimentary character of the strata from which the specimens were taken".

ORIGIN

Introductory Statement

Most of the references already mentioned include either speculations as to the origin of the deposits or brief to extensive statements of hypotheses previously proposed by others. The purpose of the following summary is to point out areas of apparent agreement or disagreement and to illustrate the applicability of mineralogical data in the continuing attempt to better define the conditions which produced the present deposits.

Source of the Tuscaloosa Sediments

There is general agreement that crystalline rocks, which prior to Upper Cretaceous time occupied the site of the present Piedmont, served as the source of the sands and clays of the Tuscaloosa formation. Assuming that these rocks were similar to those now found in the area, it appears that more than enough kaolinite could have been supplied to account for that now present in the Tuscaloosa. The A through C horizons of the present soils are estimated to contain more than 40 per cent kaolinite, vermiculite in amounts ranging from 10–40 per cent and commonly goethite, and gibbsite or montmorillonite in amounts up to 10 per cent. The quartz content ranges from 0–10 per cent (see Anonymous, 1959). If this material were to be used as a modern source of Tuscaloosa-like sediments, the quartz would either have to be (1) obtained from material below the C horizon or (2) greatly concentrated relative to the other minerals, and the abundant vermiculite would have to be (1) left behind, (2) sorted out *en route* to the depositional site or (3) altered to kaolinite during or after deposition. Unfortunately mineralogical analyses of soil profiles commonly do not include material below the C horizon and consequently to my knowledge there has been no broad, regional evaluation of the mineralogical nature of the crystalline rock in the initial weathering stages.

These points have an important bearing on the fact that although workers

have agreed on a Piedmont source, the literature records considerable disagreement as to the nature of the source material at the time of its erosion and transport to the deposition site. Readers are referred to Kesler (1956) for a review of the various hypotheses and to the original papers for the details, but, briefly stated, the contrasting points of view are presented by those of Veatch (1909) and Kesler (1951, 1956). The former postulated that uplift just before the Cretaceous resulted in erosion and removal of deep residual soils formed by weathering in effect since Silurian time. The latter pointed out that "It is now known that the region was geologically active during much of this time" (Cambrian to Cretaceous) and "Thus, any weathering of crystalline rocks of genetic importance to the Upper Cretaceous series could have occurred only briefly between Lower and Upper Cretaceous times." He believes that "the evidence favors vigorous erosion on a youthful surface, with sediment being transported directly to the ocean by numerous streams, immediately after rock disintegration and before thorough leaching".

Klinefelter *et al.* (1943) suggest that the differences of hard and soft clay types might be explained by postulating the existence of a halloysitic source for the former. Later work, to be discussed shortly, seems to provide a better explanation, and there is little evidence that differences at the source might have been maintained during transport and deposition. Furthermore, present soils and weathered rocks of the Piedmont contain intimate mixtures of kaolinite and halloysite (Anonymous, 1959; Grant, 1963; Sand and Bates, 1953) so that differences at the clay deposits would have to be explained by sorting during transportation and deposition rather than by segregation at the source.

Transportation and Deposition

There appears to be no argument that the eroded Piedmont material was transported by streams and deposited near the Cretaceous coast line in an environment characterized by anastomosing streams, oxbow lakes, and shifting deltas. Although Veatch (1909, p.100) speaks of the absence of marine conditions, he also mentions deposition of the fine clay "in the deeper and quieter waters of off-shore lakes and sounds . . . ". Subsequent workers are generally agreed that fresh water ponds, penetrated at times by the influx of salt waters, are characteristic of such environments.

Until Kesler's 1951 article appeared, most workers postulated or assumed that the clays and sands were transported from the Piedmont source to the shoreline area, sorted *en route* and at the depositional site, and emplaced in their present relative positions as part of a more or less continuous process. Kesler pointed out that the geological, mineralogical, and chemical relationships could be better explained by hypothesizing that the shore line deposits consisted of coarse feldspathic sands derived from a youthful Piedmont surface and laid down in coalescing deltas built

above sea level, "as in major deltas known today . . .". The exposed sands were then weathered and most of the feldspar altered to kaolinite prior to and during the constant reworking of the sediments by the many streams and their distributaries draining the flat-lying coastal area.

Whether the kaolinite was provided directly from the Piedmont or as a result of weathering at the deltas, all workers are agreed that the present commercial clay deposits are the result of excellent natural sorting of the fine and coarse fractions represented in the present Tuscaloosa sediments. As pointed out earlier, however, it is important to appreciate that poorly sorted clay-sand and sand-clay lenses in the Tuscaloosa are much more abundant than the commercial clays which, for good and obvious reasons, have received the greatest attention.

The manner of deposition of the fine clay sediment in the oxbow lakes and ponds of the region became an important consideration as more detailed knowledge of the composition and texture of the clay was obtained. The realization that hard clay consists of smaller particles than the soft gave rise to the initial suggestion that the two clay types were produced due to the different settling rates of coarse and fine fractions. Lack of appropriate geological evidence in support of such a hypothesis led Kesler (1956, p.553) to suggest that fresh, mildly acid water promoted "slow settling of kaolinite, with the development of platy aggregates of comparatively large size—the 'soft' kaolin", whereas upon an invasion of alkaline sea water newly inwashed kaolinite "coagulated and settled rapidly forming the dense 'hard' kaolin". Hinckley (1961), on the basis of work on flocculation by Schofield and Sampson (1954) and the textures he observed in thin section, suggested that face-to-face flocculation of clay particles in the more marine environment caused the properties responsible for a "hard" clay, whereas edge-to-face flocculation in fresh water produced the less dense soft clay.

Hinckley points out that this concept also explains many of the other mineralogical and chemical variations within and between the two types particularly if the effects of permeating solutions during and possibly after deposition of the clay are considered. He summarizes his interpretations of some of his significant findings as follows (1961, pp.127, 128):

> "The influx of the clay suspension into a saline environment resulted in a face-to-face type flocculation of clay particles which settled and carried with them the hydroxides of iron and varying amounts of boron in proportion to the pH and salinity. The resulting sediment, relatively impermeable and permitting the introduction of little dissolved material which may have been present in the surrounding solutions, compacted to a great extent with resulting high bulk density and hardness. During and after deposition recrystallization and crystal growth activity was inhibited by the lack of permeability. For similar reasons, after uplift of the deposits, relatively little oxidation and leaching of the deposits occurred.
>
> In contrast, the influx of the clay suspension into a fresh water environment was followed by an edge-to-face type flocculation of the clay particles which settled and, because of the low pH and salinity, were not accompanied by as much iron

and boron. The resulting deposit had a greater sedimentary volume, porosity, and permeability, and a lower bulk density. Its clay was relatively soft and permitted the passage of solutions containing dissolved silica and alumina, and possibly amorphous material, derived from the source area. During the depositional process, and as long as the incoming solutions were of a suitable composition, the clay particles underwent recrystallization and crystal growth. Following uplift, the greater permeability in the soft clay deposits permitted more leaching and resulted in a relative enrichment of alumina."

This picture in which salinity determined the type of flocculation which, in turn, controlled the ability of enriching or leaching solutions to permeate the clay during and after deposition, appears to satisfactorily account for the association of large particle size, good crystallinity, abundant books, higher Al_2O_3 and lower Fe_2O_3 in the soft clay, as contrasted with small particles having a lower degree of crystallinity, fewer books, lower Al_2O_3 and higher Fe_2O_3 in the hard.

CONCLUSIONS

During the past sixty-five years many workers have labored to describe measure, evaluate and explain the kaolins occurring in the Cretaceous Tuscaloosa formation in the Southeastern United States. All have contributed in one way or another to our understanding of the important commercial deposits of the area. Detailed studies of the clay mineralogy during the past twenty years have done much to account for some of the variations in the properties and behavior of the clays. However, in the light of the ever-increasing need for clay and clay products possessing specific characteristics necessary to meet the demands of a booming technology, much careful, detailed geologic and mineralogical work remains to be done. Particularly useful in filling some of the gaps in our present knowledge will be future studies of Tuscaloosa clays and sands properly sampled at varying distances from commercial pits; for it is only in this way that a more representative picture will be obtained of the entire Tuscaloosa formation, and of the relationships of the commercial clays of today to those certain to be required tomorrow.

Finally, I would like to point out that there is a great deal of information on these clays which, for reasons probably well justified in the past, has not found its way from company files into the literature. With the recent increased development of well-equipped research laboratories in the area, it is to be hoped that the reviewer twenty years hence will be able to discuss many basic contributions by the company scientists and engineers who are and will be doing most of the work in the region.

ACKNOWLEDGMENTS

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