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MINERALOGICAL AND CHEMICAL VARIATIONS IN THE KAOLIN DEPOSITS OF THE COASTAL PLAIN OF GEORGIA AND SOUTH CAROLINA

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ABSTRACT

Three hundred forty-three samples from so-called hard and soft kaolin deposits in the Atlantic Coastal Plain of Georgia and South Carolina were evaluated in terms of twenty-two chemical and mineralogical properties.

The purpose of this investigation was to obtain a better understanding of the origin and variability of the deposits. Samples were taken largely according to pre-determined sampling plans formulated to answer the questions, a) are the deposits homogeneous in terms of the measured properties and b) can the hard and the soft types be distinguished on the basis of these properties?

All variables tested by statistical methods are shown to be non-homogeneously distributed within the deposits. The hard and soft clay types can be distinguished by an analysis of variance in terms of the variables Al_2O_3, Fe_2O_3 and kaolinite crystallinity; and possibly by several other variables which, although not shown to be statistically different, are strongly characteristic of one of the types. Al_2O_3 and crystallinity have greater values in the soft, and Fe_2O_3 has consistently higher values in the hard type. The larger particle sizes are associated with the soft type, and microscopic examination of thin sections shows that the hard type has feathery-patches of parallel-aligned flakes, in contrast to books interspersed in an isotropic matrix for the soft type.

In the soft type, positive correlations between Fe_2O_3 and mica, Fe_2O_3 and K_2O, K_2O and mica, and the negative correlations between mica and books, and Fe_2O_3 and books, suggest that this type of clay may have been affected by leaching processes.

A possible explanation of the differences observed in the hard and soft kaolin deposits is that they were deposited in saline versus fresh water environments, respectively. The influx of a clay suspension into a saline environment resulted in a face-to-face type flocculation with attendant higher compaction, less permeability, and because of higher pH, more hydroxides of iron than would result from a fresh water environment. In the hard type the lack of permeability was because of relatively less recrystallization and crystal growth activity during and shortly after deposition. Similarly, this would account for less leaching action following uplift than was able to occur in the more permeable soft type.

INTRODUCTION

The kaolin deposits investigated occur along the "fall line" in the Coastal Plain province of Georgia and South Carolina and are contained in the Tuscaloosa formation of Upper Cretaceous age (Cooke, 1943). They have no preferred stratigraphic position in this formation and are generally lenticular in shape, sometimes attaining a thickness of 50 feet and a maximum horizontal dimension of perhaps a mile. They usually have gradational contacts with the enclosing sediments which in this area

[1] Deceased.

consist of an unconsolidated series of micaceous, kaolinitic, fine to very coarse sands (Kesler, 1956).

Differences among the deposits, first pointed out by G. E. Ladd in 1898, led early investigators to classify them into hard, semi-hard and soft varieties. A deposit may sometimes consist of a layer of hard clay overlying a soft layer, but other than this there is generally no sharp stratification within the lens (Kesler, 1956).

The purpose of this investigation was to obtain a better understanding of the origin and the variability of the deposits by a study of data from selected chemical and mineralogical properties. A total of three hundred forty three (343) samples, obtained from thirty two surface exposures and nine pairs of drill cores from both hard and soft type clay deposits, were analyzed.

Previous Studies

Variations among the clay deposits were noticed by early investigators (Ladd, 1898; Veatch, 1908; Shearer, 1917) and many of the physical properties affecting economic utilization of the clays were recorded by them along with extensive geological descriptions. Later, investigators Stull and Bole (1926) classified the clays into flint, hard, semi-hard, soft and bauxitic types. Neumann (1927), made no distinction among the South Carolina deposits as to hardness or type but presented evidence related to the cause of the whiteness of the deposits. Smith (1929) described many clay localities and presented numerous chemical analyses.

The work of Klinefelter *et al.* (1943) was a singular contribution toward establishing chemical and mineralogical differences among the clays and in relating these differences to variations in hardness. In a concurrent but independent study, Mitchell and Henry (1943) related hardness and certain mineralogical properties to specific behavioral properties. A field investigation by Kesler (1956) provided additional information and insight on possible mechanisms of deposition and modes of origin, and pointed out the need for detailed mineralogic work. The present article summarizes the findings detailed in a Ph.D. thesis (Hinckley, 1961) and in part reported on in Hinckley and Bates (1960), Hinckley (1963) and Bates (1964).

Theories on Origin of the Deposits

Theories on the origin of the deposits are conveniently presented in three parts; the source area, the depositional site, and the nature of post depositional changes.

Both Veatch (1908), and Neumann (1937), proposed the source of the sediments to be a thoroughly rotted Piedmont area to the northwest, containing a red surficial covering over-lying a gray kaolinitic mass of rock weathered to considerable depth. Uplift in this area caused a flood of sediment southeastward toward the Cretaceous sea. Kesler (1956), on the other hand, visualized a youthful Piedmont source area undergoing vigorous erosion, but with sufficient weathering and leaching to decompose and remove the mafic minerals. Slight reworkings of the resulting feldspathic delta and flood plain material, after additional weathering, allowed the kaolinite to be separated from the sands and to be deposited in cut-off river segments and ponds.

Both Veatch (1908) and Kesler (1956), as well as others (Berry, 1914) stressed the non-marine conditions attending the environment of deposition and both included the possible differing influence of saline and river water, although Kesler related salinity specifically to hardness. Smith (1929) suggested that differences in hardness reflect differences in the depositional site and that hard and soft clays may have been deposited continuously from the same source, the coarser particles settling first, forming the soft clays, and the finer particles settling later, forming the hard clays.

In an early investigation (Shearer, 1917) it was proposed that hydrogen sulfide bearing springs had existed within the depositional basin and were the cause of the nodules of iron sulfide and pockets of bauxite. Kesler (1956) considered that the oxidation of sulfides, and Stull and Bole (1926, p. 6) that the leaching and weathering of the deposits, largely contributed to the occurrence of bauxite. Stull and Bole (1926) suggested that hardness was imparted by free silicic acid and that leaching of silicic acid from the hard clay formed the soft. Henry and Vaughan (1937) conversely suggested that infiltration of silicic acid from overlying fuller's earth changed the soft to the hard type. Smith (1929) earlier suggested that all the clay may have been deposited as the soft type but that the upper part of certain deposits was changed to hard type by some process of alteration similar, possibly, to the formation of bauxite by lateritic weathering.

It is left to the reader to pursue in the original articles the interesting ramifications of these and other theories proposed by the earlier investigators.

Experimental Methods

Sampling. Samples were taken according to several predetermined sampling plans, but only the principal sampling plan is described in detail here; the others, described in detail in the thesis (Hinckley, 1961), are of a similar type and their results generally only serve to corroborate the information obtained. Two questions formed the basis for this sampling plan and its associated experimental design: (1) are the deposits homogeneous in terms of certain measured properties? (2) can the hard and soft clay types be distinguished on the basis of these properties?

The principal sampling plan utilized eight drill cores, taken in pairs from four commercially active clay deposits, two hard type and two soft type. The clay deposits were designated by the company supplying the core as hard or soft, even though some differences were known to exist among the cores from the same deposit. To insure overall coverage, the sampling plan, a nested type (Bennett and Franklin, 1954) required that each core be arbitrarily divided into thirds and that two six-inch samples be taken at random from each third. Each six-inch sample was then divided into three two-inch subsamples providing eighteen subsamples from each core, or a total of 144 two-inch subsamples from the eight cores.

Variables and Analytical Methods. Twenty-two properties were measured on varying numbers of samples and, together with the analytical methods used, are listed on Table 1. A complete description of the analytical techniques used and the assumptions made is not presented here but can be found in the thesis (Hinckley, 1961).

Presentation and Analysis of Data

Analysis of variance and simple correlation were the principal statistical methods used in the data analysis. The data from which these statistical analyses were made are not included in this report but are to be found elsewhere (Hinckley, 1961). The summary statistics for the data and tables summarizing the results of the data analyses are included, however, and appear in the following sections according to the method of data analysis used.

Analysis of Variance. All variables tested by the analysis of variance are shown to be non-homogeneously distributed within the deposits. The word non-homogeneous is used in a relative sense, usually comparing the variation at a particular sampling level to the variation occurring among subsamples, or in some cases to a pooled error term (Bennett and Franklin, 1954, p. 408). The summary statistics for all the variables quantitatively measured appear on Table 2. Table 3 contains the data for ten of the variables, grouped according to clay pits. On the basis of the variability occurring within the clay deposits, the difference between the hard and soft type clay deposits is shown to be significant at the 95% probability level in terms of Al_2O_3, Fe_2O_3 and crystallinity. The Al_2O_3 and crystallinity values are higher in the soft type while in the hard type the Fe_2O_3 content is higher. Larger particle size, as measured on electron micrographs from eight samples, is shown to be associated with the soft type by a Chi square test. In addition, a number of other variables also contrast the hard and the soft types, although not shown to be statistically significant principally because of the small number of samples studied. Higher boron and bulk density values, greater resistance to dispersion and a contorted "worm burrow" type texture tend to be associated with the hard type. More abundant books, and graphic and patchy type textures characterize the soft type.

TABLE 1. MEASURED VARIABLES AND METHOD OF ANALYSIS

Variables	Method of Analysis
SiO_2, Al_2O_3, Fe_2O_3, TiO_2, MnO, K_2O	Fluorescent x-ray spectroscopy (Birks, 1959)
Montmorillonite	Fluorescent x-ray spectroscopy in terms of exchanged strontium (Hinckley and Bates, 1960)
Crystallinity	Degree of crystal perfection of Kaolinite as determined from x-ray diffractometer tracings (Bates and Hinckley, 1959; Hinckley, 1963)
Quartz, mica, kaolinite books, undispersed clay particles >200 mesh	Point count methods under the petrographic microscope (Griffiths, 1952)
B, Ga	Emission spectroscopy (O'Neil and Suhr, 1960)
Bulk density	Mercury immersion balance (Rosenfeld, in Griffiths, 1952)
Ca, K (Exchanged)	Flame photometry (O'Neil, 1958)
Particle size	Electron microscopy (Hinckley, 1961, p. 47)
Thin section examination of microstructure. Texture, roughness, hardness evaluation	Light microscopy and megascopic observations

A summary of the analyses indicating the variability existing at different sampling levels, and the results of the "F" tests (Snedecor, 1959, p. 244) appear in Table 4.

Simple Correlation. The simple correlation coefficient was used to estimate the degree of association between pairs of selected variables. The

Table 2. Summary Statistics According to Clay Type

Variable	Clay Type	Mean	Standard Deviation	Range	Sample Size
SiO_2	Hard	44.1%	.49	43.2–45.5	50
	Soft	43.8%	.75	41.1–47.9	197
Al_2O_3	Hard	38.6%	1.07	35.4–40.7	50
	Soft	39.7%	1.26	31.9–42.3	197
K_2O	Hard	.28%	.19	0.02–0.76	50
	Soft	.14%	.14	0.00–1.12	197
Fe_2O_3	Hard	1.9%	.71	0.75–5.52	50
	Soft	.2%	.23	0.00–1.64	197
TiO_2	Hard	1.6%	.27	1.09–2.12	50
	Soft	1.5%	.48	0.43–3.87	197
Montmorillonite	Hard	2.3%	.78	1.00–3.46	50
	Soft	3.0%	6.86	0.01–50.30	197
Crystallinity	Hard	.46[1]	.13	0.28–0.75	50
	Soft	.90[1]	.22	0.55–1.43	197
>200 mesh	Hard	2.13%	1.71	.05–6.54	50
	Soft	.67%	.69	.04–2.73	197
Quartz	Hard	69.0[2]	65.0	2–189	50
	Soft	4.0[2]	9.0	0–64	197
Mica	Hard	38.0[2]	58.0	0–171	50
	Soft	94.0[2]	60.0	0–85	197
Books	Hard	2.0[2]	3.0	0–12	50
	Soft	75.0[2]	50.0	13–190	197
Undispersed clay	Hard	64.11[2]	75.82	0–196	36
	Soft	14.23[2]	20.82	0–100	72
Boron	Hard	1.06[3]	.099	.91–1.16	8
	Soft	0.65[3]	.264	.40–.96	8
Gallium	Hard	1.62[3]	.099	1.47–1.75	8
	Soft	1.68[3]	.063	1.61–1.78	8
MnO	Hard	359[4]	150	174–697	12
	Soft	356[4]	98	251–613	12
Bulk Density	Hard	1.620	.024	1.514–1.734	12
	Soft	1.480	.016	1.454–1.505	12
Exchangeable Calcium	Hard	72.5[5]	2.60	70–75	12
	Soft	120.8[5]	45.81	65–190	12
Exchangeable Potassium	Hard	31.8[5]	2.30	29–36	12
	Soft	33.0[5]	2.41	29–36	12
SiO_2/Al_2O_3	Hard	1.16	.05	1.07–1.29	72
	Soft	1.11	.03	1.06–1.22	72

[1] Index proportional to degree of crystal perfection.
[2] Grains per 200 points in thin section.
[3] Reciprocal % transmission×10 (emission spectroscopy).
[4] Counts per 40 seconds (fluorescent x-ray spectroscopy).
[5] Parts per million.

coefficients obtained in this study were tested for significance by use of tables of "r" found in Arkin and Colton (1950, p. 140). It should be mentioned that while these coefficients reflect the relationship between pairs of variables, the interpretation may change when other inter-dependent variables are taken into account.

In order to enhance the indication of possible relationships existing among the variables, independent of the coarser detrital grains, the data

TABLE 3. SUMMARY STATISTICS ACCORDING TO PITS

Pit Number	Sample Size	SiO_2	Al_2O_3	K_2O	Fe_2O_3	TiO_2	Montmorillonite	Crystallinity Index	Quartz	Mica	Books
					Mean Value						
1	36	43.76%	39.90%	.10%	.17%	1.53%	1.39%	.94[2]	17.6[3]	65.6[3]	100.0[3]
2	36	43.99	39.52	.20	.27	1.54	2.60	.82	1.1	107.2	71.6
3	24	43.82	39.69	.16	.51	1.44	1.71	.90	5.5	132.3	53.2
4[1]	36	44.80	38.19	.50	1.91	1.65	1.96	.51	42.9	78.7	1.3
5	24	43.75	39.68	.24	.25	1.67	1.91	.88	5.8	119.1	63.5
6	24	43.68	39.62	.26	.31	1.48	6.52	.88	1.0	112.2	64.5
7	24	43.78	39.72	.28	.49	1.48	2.09	.89	4.7	126.9	42.6
8	24	44.77	38.49	.23	.36	1.80	1.45	.73	49.7	98.5	18.9
9[1]	36	43.82	38.43	.18	1.84	1.66	2.64	.40	126.2	.8	.2
					Standard Deviation						
1	36	.44	.86	.06	.14	.52	1.08	.20	3.48	60.73	53.43
2	36	.40	1.23	.13	.19	.54	1.86	.13	2.16	55.41	54.56
3	24	.29	.51	.19	.57	.20	.99	.20	12.70	26.49	23.79
4[1]	36	.87	1.27	.31	.84	.21	.63	.12	48.70	58.49	3.65
5	24	.62	.81	.33	.26	.50	.86	.21	10.16	41.32	35.35
6	24	1.14	1.39	.38	.30	.33	14.00	.17	.41	66.26	55.27
7	24	.43	.74	.25	.37	.21	.92	.23	14.06	62.44	27.43
8	24	1.82	1.47	.27	.38	.40	.47	.09	18.68	26.56	7.40
9	36	.34	1.06	.09	.41	1.02	.86	.09	53.56	1.40	.48

[1] Hard type clay.
[2] Index proportional to crystal perfection.
[3] Grains per 200 points in thin section.

for samples that contained abundant grit, as determined in the texture analysis, were separated. After this separation the remaining data were analyzed as hard and soft types and then as a combined group. Table 5 comprises the resulting correlation coefficients and indicates by an asterisk the stronger correlations.

In addition to the correlations recorded on Table 5, several strong associations are indicated by a Chi square test for independence (Bennett and Franklin, 1954), among the less quantitatively measured variables. Larger clay particle size and graphic and patchy textures (Fig. 1), for example, are associated with the soft type, and there is evidence that samples showing high test-hardness are associated with a smooth feel, a

TABLE 4. VARIATION IN CHEMICAL AND MINERALOGICAL PROPERTIES

Source of Variation	Degrees of Freedom		Variation (Mean Square Value)										
			SiO_2	Al_2O_3	K_2O	Fe_2O_3	TiO_2	Montmor-illonite	Crystal-linity	Quartz	Mica	Books	SiO_2/Al_2O_3
Clay Types	1	Combined	6.25	70.56*	10,677	953,552**	4,888	101,177	73,486*	170,100	25,643	218,599	97.2
	1	Hard[1]	19.81	.05	18,432	1,371	561	73,408	1,922**				
Pits	1	Soft[2]	.31	1.42		983	227	513,084	5,495***				
	2	Combined[3]	8.61	.73	9,609	1,150	394	293,246	3,708**	41,169**	66,524**	17,454*	7.8
	2	Hard	5.28*	2.73	6,305*	6,422	10,279	61,537*	263.5				
Cores	2	Soft	.43	4.33		62	4,600	374,262 *	181.7				
	4	Combined	3.58*	3.54	3,198**	3,256	7,440	217,900**	222.9	7,264	5,145	8,152	9.6
	8	Hard	.43	4.29	1,226	21,917**	25,448	16,861	316.0				
Thirds	8	Soft	.26	3.00**		1,130	7,562	48,128	965.6				
	16	Combined	.53	3.77*	863	11,523***	16,505	32,620	640.8	3,365	6,829*	5,012	4.3
	12	Hard	1.01**	2.53**	1,132***	2,062	16,012***	12,971***	146.4*				
Samples	12	Soft	.26	.79		642***	7,438***	28,239***	551.1***				
	24	Combined	.75***	1.58*	652***	1,352	11,727***	20,521***	348.7***	12,476	2,570**	4,521*	2.6
	48	Hard	.07	.72	23	2,147	37	212	63.3				
Subsamples	48	Soft	.17	1.11		55	1,596	322	75.5				
	96	Combined	.15	.92	15	1,101	817	267	69.4	7,883	310	2,515	.73

*, **, ***, the "F" ratio formed using this figure and the error term as noted in text, is significant at the 5%, 1% and .1% level respectively.

[1] Sample size of hard type = 72.

[2] Sample size of soft type = 72.

[3] Sample size of combined hard and soft types = 144.

TABLE 5. SIMPLE CORRELATION COEFFICIENTS

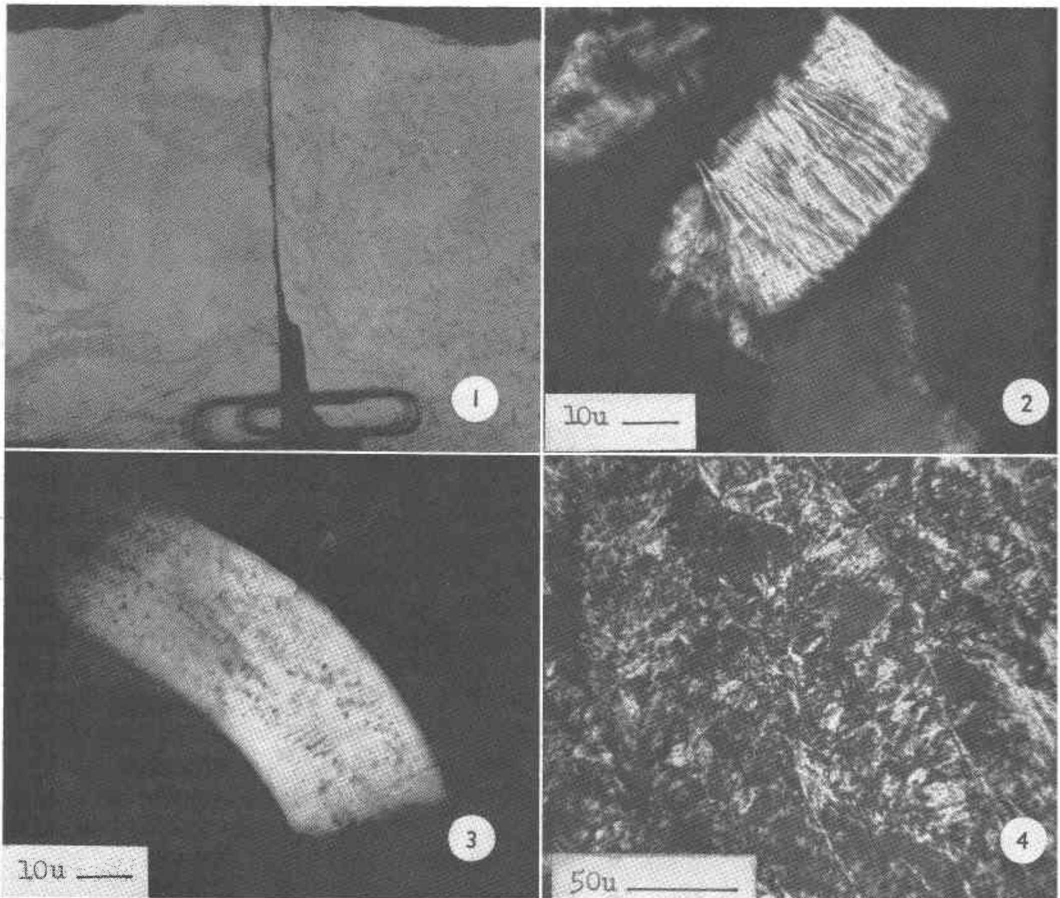
	SiO_2	Al_2O_3	K_2O	Fe_2O_3	TiO_2	Montmorillonite	Crystallinity	Quartz	Mica	Kaolinite Books	Boron[4]
	—[1]	−.11	.43*	−.14	.32	−.32	.37	−.16	.42*	.14	
SiO_2	—[2]	−.47*	−.02	−.09	−.26	.51*	−.01	−.01	−.12	−.08	
	—[3]	−.44*	.10	.07	−.21	.47*	−.07	.05	−.09	−.13	.45
			−.52*	−.47*	.07	−.01	−.21	.07	−.37	−.31	
Al_2O_3			−.07	−.06	−.19	−.62*	.21	.05	.05	.12	
			−.28	−.39	−.15	−.52*	.34	−.20	.10	.28	−.71*
				.44*	.05	−.11	.45*	−.21	.80*	.48*	
K_2O				.61*	.08	−.07	−.22	.08	.26	−.31	
				.54	.07	−.07	−.31	.16	.28	−.39	.68*
					−.48*	.30	−.08	.01	.35	.43*	
Fe_2O_3					−.10	−.06	−.33	.01	.51*	−.45*	
					−.08	−.04	−.65*	.57*	−.13	−.57*	.75*
						−.82*	.57*	−.48*	.15	.08	
TiO_2						−.15	.15	−.08	−.15	.24	
						−.16	.14	−.11	−.10	.19	.40
							−.56*	.59*	−.38	−.39	
Montmor-							−.32	−.07	−.09	−.24	
illonite							−.21	−.02	−.07	−.18	.73*
								.27	.40*	.33	
Crystal-								.19	−.55	.47*	
linity								−.45*	−.06	.65*	−.76*
									−.50*	−.38	
Quartz									−.08	−.11	
									−.39	−.39	.65*
										.73*	
Mica										−.79*	
										−.35	−.07
Books											−.94*

[1] (Upper), hard type N = 50.
[2] (Middle), soft type N = 197.
[3] (Lower), combined hard and soft type N = 247.
[4] Boron group, combined hard and soft type N = 16.
* Significant at least at 95% level.

textureless appearance and a high montmorillonite content (Hinckley, 1961).

Thin Section Observations. A study was made by use of the petrographic microscope of forty-one thin sections from samples taken by a random process from cores from four deposits and from selected samples obtained from several other deposits.

Kaolinite books are observed in abundance in thin sections of the soft



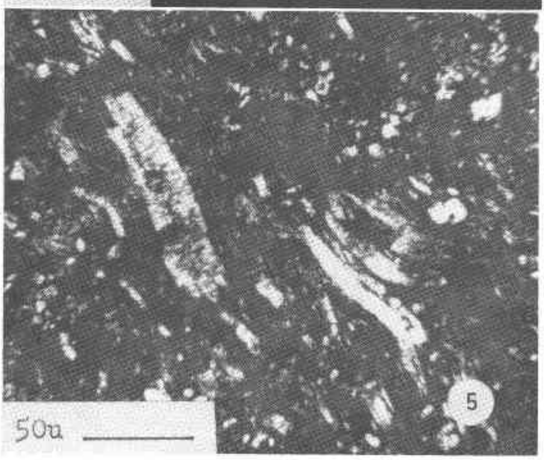


FIG. 1. Photograph of patchy texture (left half) and graphic texture (right half). These textures, and varieties thereof, are commonly observed in the soft type clays. The patchy texture commonly shows light patches in a darker matrix but may show the reverse. Coarse graphic texture grades into fine patchy texture.

FIG. 2. Photomicrograph of a kaolinite book. The "shaggy" appearance is characteristic of books newly formed from mica weathering in situ.

(Balance of legend appears on the facing page)

type clay, and their relatively strong birefringence and distinctive size and shape set them apart from the surrounding clay matrix. They are not commonly observed in the hard type, and when observed they are generally not large or well developed. The shape of the books occurring in the soft clay, although occasionally long and sinuous, is more often prismatic. In comparison to the books observed to be forming in situ from the weathering of mica in the Piedmont and occasionally in the Coastal Plain deposits, which are feathery-edged and irregular, the edges and surfaces of these books are characteristically much smoother and better defined. This difference is illustrated on Figs. 2 and 3.

Striking contrasts exist in the hard and soft types of clay with respect to the birefringence patterns observed in thin sections. The soft type is characterized by discrete, relatively high-birefringent books of varying size which are dispersed in a largely isotropic clay matrix. In contrast the hard type rarely shows the relatively high-birefringent books. It is characterized by irregular patches and feather shaped aggregates of parallel-aligned particles of relatively moderate birefringence alternating with irregular isotropic areas. The appearance of these birefringence patterns is shown in Figs. 4 and 5.

Mica, in the process of weathering to kaolinite, is observed in both the hard and soft type deposits. The weathered condition of the mica ranges from a slightly lower-birefringent edge on an otherwise fresh mica flake to what appear to be leached clumps of irregular, feathery-edged but distinct kaolinite books. The soft type has a greater proportion of mica flakes and shows more abundant weathered particles. Although no count data were taken, the proportion of weathered flakes appeared about equal in both types.

Scattered "mica-like" flakes of material, probably similar to those reported by Ross and Kerr (1930, p. 172) and characterized by a birefringence similar to that of muscovite, but with distinctly lower indices of refraction, were also seen in both types.

FIG. 3. Photomicrograph of a kaolinite book. The "trim" appearance is characteristic of a large majority of the books occurring in the soft type clay.

FIG. 4. Photomicrograph of a typical birefringence pattern obtained in thin section from a hard type clay. The white, "whispy," "feather-shaped" patches and long stringers are caused by a parallel alignment cf clay particles. Such parallelism is indicative of face-to-face flocculation.

FIG. 5. Photomicrograph of a typical birefringence pattern obtained in thin section from a soft type clay. The white, angular, usually elongate patches are books. The largely isotropic area between the books indicates a non parallel orientation of the smaller particles.

Discussion of the Results

Variability of the Deposits. The experimental design used in this investigation was formulated to answer two specific questions: (1) are the deposits homogeneous with respect to particular measured properties? (2) can the hard and soft clay types be distinguished on the basis of these properties?

Without exception, the fourteen variables tested by analysis of variance methods were shown to be nonhomogeneously distributed within the deposits.

On the basis of the principle sampling plan it is not possible to determine the actual distribution of the variables within the deposits. It is evident, however, that the values for the different properties do not closely parallel each other and that the variation within a six-inch sample is generally less than between these six-inch core samples. This general pattern consistent over all variables tested and over both types has two interesting exceptions; the Fe_2O_3 in the hard type and the Al_2O_3 in the soft type, both appear to be varying in broader units, thirds of cores. (See Table 4.) For the most part such units could not be distinguished megascopically. Extra samples from pairs of cores from two soft type clay deposits were analyzed, however (Hinckley, 1961), in an attempt to detect a correspondence with megascopic features and possibly a layering extending between cores. Of the variables analyzed, quartz, books, Fe_2O_3, crystallinity and montmorillonite, only in the case of montmorillonite was there a correspondence with the megascopic zones and evidence for possible layering in both pits. Fe_2O_3 and crystallinity values coincided with the same megascopic zones, but satisfied the test for consistency between cores in only one pit.

Even though the hard and the soft type clays may be distinguished, and in the case of Fe_2O_3, Al_2O_3, crystallinity and particle size on a statistically significant basis, the question as to whether the deposits so distinguished are end members of a single population, or whether they represent two distinct populations, cannot be determined from these data. However, the evidence obtained in this investigation points to the existence of two genetically distinct populations whose principal differences are directly or indirectly related to depositional conditions which controlled the type of flocculation of the influxing clay suspensions.

Origin of the Deposits

1. *Source Material*

The source material from which the kaolin was derived is generally agreed by most investigators to have been a crystalline Piedmont area to the northwest, but the extent to which it was weathered and the possible mechanisms of Nature's "clay washing plant" are not agreed upon. It is also generally agreed that no apparent difference exists between the two clay types insofar as preferred geographic setting is concerned, with the exception that the hard type may be found more often above the soft when the two are found together (Stull and Bole, 1926); but as yet, a satisfactory explanation of the present differences between the hard and the soft, on the basis of different types of source material, has not been proposed. From the available evidence it appears probable that within rather narrow limits the source material for the two types was the same.

2. *Depositional Environment*

Differences among the deposits probably arise, in part, directly from differences in the depositional environment, particularly the salinity, pH, and motion of the water and, in part, from secondary or post depositional processes.

The effect of the motion of the water in the basin of deposition, within the limits generally proscribed for the sedimentation of clays, may not have the influence attributed to it by Smith (1929) for the separation of the hard and soft types by differential settling. According to Whitehouse and Jeffrey (1958, p. 56), stratification within clay deposits " . . . does not arise or persist as a consequence of differential settling. Such stratification can only occur as the result of definite environmental changes." Variations in turbulence and velocity of the water, particularly near shore, would be expected to have an effect on the amount of grit, initial textural features, and perhaps on particle orientation.

The type of flocculation may have affected the rate of deposition and thus influenced the textural appearance and possibly the grit content of the deposits. Recent work by Whitehouse and Jeffrey (1958) on differential settling tendencies of clay minerals suggests that the nature of the settling unit (the floc), which controls the rate of settling, could have influenced the ability of the clay water system to transport the large detrital grains into what appears to be an incongruous depositional environment. No attempt was made in this investigation to consider possible hydraulic equivalence relationships among flocs and detrital grains.

The contrasting birefringence patterns, independent of the kaolinite books, indicate that different flocculating conditions existed at the time of deposition. The isotropic background in the soft type clay indicates a lack of parallel particle orientation as would be expected in a face-to-edge type flocculation. The parallel-aligned particles forming anisotropic patches in the hard type clay indicate a face-to-face type flocculation. Without exception in the 41 thin sections examined the face-to-face type birefringence pattern occurred only in the hard type.

These two flocculation types have been described by Schofield and Sampson (1954), and related to the salt content of the clay suspensions. The edge-to-face type occurs in salt-free suspensions and flocculates with a large sedimentary volume, while the face-to-face type occurs with high salt concentrations and results in a smaller sedimentary volume. The conditions which lead to a particular type of flocculation may vary and " . . . will depend both on the concentration of the clay and the concentration and kind of salts" (White, 1961, p. 566).

The removal of water from a face-to-face flocculated clay " . . . leads to the development of capillary forces within the mass whose magnitude increases with decreasing pore diameter . . . " (Michaels, 1958, p. 27). Such a genetic process in the case of the hard clay type, in contrast to the soft, would tend to give it greater bulk density and because of the greater number of interparticle contacts, generally greater dry strength. In this investigation the mean difference in bulk density found between the hard (mean density = 1.62) and the soft (mean density = 1.48) is not statistically significant for the few samples (24) on which this variable was measured, but there is no overlap in values, the hard type being consistently more dense than the soft.

The ease with which the clays can be dispersed in water shows considerable variation within clay types and distinct differences between clay types (Stull and Bole, 1926). This difference was also found in this investigation and the large number of undispersable clay aggregates from the hard type, about four times more abundant than from the soft, is also in agreement with the data given by Klinefelter *et al.* (1943, p. 11). The reason for this difference in susceptibility to dispersion was considered by these investigators to be largely a matter of grain size, the hard type postulated to be a smaller particle size. The work of Michaels (1958), however, indicates that particle orientation and packing may be of more importance to dispersion than particle size, and the investigation of Whitehouse and Jeffrey (1958), provides a possible explanation for the resistance of the hard type to dispersion, in terms of genesis.

"Clay materials that have been exposed to ocean water exhibit a dispersal resistance to ultimate deflocculation that is distinctly different from the resistance displayed by similar materials not so exposed." (Whitehouse and Jeffrey, 1958, p. 56).

If saline water were the cause of the face-to-face flocculation of the hard type, assuming a uniform and continuous source material, the work of Landergren (1945) as reported by Goldschmidt (1954, p. 285) suggests that the boron content should be higher in the hard type. The results of this investigation indicate that the mean difference in boron content between the hard (32 ppm) and the soft (1 ppm) is not statistically significant for the 16 samples analyzed, but that the hard type does contain the greater amount with but one exception.

The higher pH of saline water would have caused a greater amount of iron to precipitate and would account for the significantly greater Fe_2O_3 content of the hard type. The positive correlation between Fe_2O_3 and boron (.75) would also be expected if the cause of the face-to-face flocculation were saline waters.

3. *Post Depositional Changes*

Post depositional changes in the deposits were probably profoundly affected by the type of flocculation. The susceptibility of a deposit to leaching would depend to a large extent on its permeability which, according to filter pressing tests, is strongly influenced by the type of flocculation. The edge-to-face type flocculation with " . . . relatively high void volume and large void space results in both rapid filtration or drainage and rapid drying" (Michaels, 1958, p. 27).

The reported occurrence of bauxite within a few soft type deposits is suggestive of leaching action, possibly involving the oxidation of pyrite as described by Kesler (1956). He supports this observation by pointing out that the occurrence of pyrite is largely restricted to the hard clay; and the data on Fe_2O_3 presented herein—with means of 1.9% in the hard clay and 0.2% in the soft—may be taken as a further indication of such a possibility.

In the samples measured, the SiO_2/Al_2O_3 ratio is below the theoretical value of 1.18 for kaolinite by about one per cent in the hard and 2.5 per cent in the soft clays.

The presence of quartz as detected in 16% of the *x*-ray patterns of 50 samples of hard clay and 2% of the patterns of 197 soft clay samples may help explain part or all of this difference between the two clay types, but, when considered in the light of the low SiO_2/Al_2O_3 ratio relative to kaolinite, it also suggests that excess alumina may be present in a significant proportion of the samples. A careful examination of the *x*-ray patterns from the soft type core samples did not reveal the presence of gibbsite, suggesting that, at least in the samples studied, any excess alumina is present in a form or amount undetectable by the *x*-ray procedure used. Examination of the Al_2O_3 values shows no preferential distribution of alumina within the cores.

The differences between clay types in pyrite content, the SiO_2/Al_2O_3 ratio, and the amounts of Fe_2O_3 and Al_2O_3 support the contention that the soft type deposits have been subjected to greater leaching and oxidation than the hard type. The lack of a correlation between Fe_2O_3 and Al_2O_3 in the soft clays indicates, however, that this leaching has not been of the prolonged lateritic type in which the hydroxides of iron and aluminum have accumulated together.

Similar conclusions can be inferred from the fact that TiO_2 is not significantly correlated with Al_2O_3 in the samples studied. However, lack of data as to the relative abundance of rutile and anatase limits the usefulness of this variable in distinguishing possible differences within and between deposits.

The type of flocculation and its effect on permeability is probably the underlying factor causing the differences in crystallinity values between the clay types. Klinefelter *et al.* (1943) first noticed the difference and suggested that it was probably due to some underlying common factor which also caused hardness. The uniformity of the low crystallinity index values in the hard type, in contrast to the large variations in the soft, may also suggest more uniform depositional or post depositional conditions.

The generally clean cut appearance of the kaolinite books found in these deposits, regardless of size or curvature, suggests that either solution, or possibly abrasive action, has removed the irregular, feathery edges that are commonly found on books forming directly from mica, or that they are authigenic in origin. The presence of the long sinuous forms and the parallel and often intricate association of numbers of small books is moreover evidence for authigenic development and the abundance of these books of all sizes in the soft clay, and their paucity in the hard, suggest that conditions for crystal growth were not favorable in the hard type. The strong positive correlation between crystallinity and books in the soft type suggests that these two variables responded together to a recrystallization, crystal growth process; and the abundance of authigenic kaolinite books would contribute to the negative correlation between mica and books. Recrystallization and crystal growth was probably enhanced in the soft type by the edge-to-face flocculation inasmuch as it would increase the available surface area and the permeability of the clay deposits.

The classification of the kaolin deposits into hard, semi-hard and soft types principally on the basis of the ease with which the air-dried raw kaolin sample could be pulverized in the hand, led Klinefelter *et al.* (1943), at the conclusion of their excellent work in which they measured a number of properties on nineteen clay samples of varying hardness, to state that, "Although the clays have been divided into three groups—soft, semi-hard, and hard—in most of the tests the semi-hard clays reacted like the hard varieties."

It appears from the available evidence that while on the basis of "hardness" or pulverizing ease, as it may be affected by flocculation type, montmorillonite content, and particle size, there may be a single population with hard and soft end members, the distribution of other properties suggests the existence of two genetically distinct populations.

As a possible interpretation of some of these differences, it is suggested that for a fairly uniform source material, the differences are probably closely related to a saline versus a fresh water depositional environment as follows.

The influx of the clay suspension into a saline environment was followed by 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 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, subsequent to the reduction of the iron and its formation into pyrite, relatively little oxidation and leaching of the deposits occurred.

In contrast, in the soft clay situation, an influx of the clay suspension into a fresh water environment was followed by an edge-to-face type flocculation of 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, 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, resulting in improved crystallinity, increased particle size, and the growth of large numbers of kaolinite books. Subsequent to uplift of the deposits the environment changed to one of oxidation, as in the case of the hard type, but because of the greater permeability in the soft clay deposits, leaching was able to occur to a greater extent (the degree depending on the local topography), resulting in a deficiency of silica.

The superposition of the hard type on the soft, when the two are found together, may be due to a process involving a gradual subsidence of the source area, with a corresponding transgression of the sea which tended to change fresh water depositional environments to saline water more often than the reverse.

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