ON THE GENESIS OF RESIDUAL KAOLINS*

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ABSTRACT

A study of weathered feldspathic rocks in the Southern Appalachian region revealed that hydrated halloysite forms by the weathering of all types of feldspar where environmental conditions are favorable, as in the Spruce Pine district of western North Carolina. Further south and north of this district in the Blue Ridge province and east into the Piedmont province, the potash feldspar increasingly alters to vermicular kaolinite through the intermediate stage of secondary mica, until all feldspars alter to kaolinite and only small amounts of hydrated halloysite are formed. Hydrated halloysite, and not its dehydrated product, is found in all deposits, which indicates environmental conditions have not changed appreciably since Tertiary time. Hydrated halloysite and kaolinite are formed independently; there was no evidence of transitions between the varieties. Primary mica and secondary mica formed by weathering of feldspar always alter to vermicular kaolinite.

Quantitative mineralogical analyses were made on samples from most of the deposits by the thin-section point counter technique and by *x*-ray diffraction and glycol-differential thermal analyses of size fractions.

INTRODUCTION

Kaolinite and hydrated halloysite occur as mixtures in residual clay deposits. Halloysite occurs as a dehydration product in some deposits in exposed mine faces. The reason for the occurrence of mixtures of kaolinite and hydrated halloysite had not been established clearly. For this reason a detailed mineralogical study of the residual kaolins of the Southern Appalachian region was undertaken in an attempt to ascertain the conditions giving rise to these compositionally similar clay minerals during the processes of weathering. In both the Blue Ridge and Piedmont physiographic provinces, good deposits for sampling are afforded by the strip mines producing mica and kaolin derived from the weathering of granites and pegmatites. An areal map showing distribution of the areas sampled is shown in Fig. 1. The deposits sampled in western North Carolina are described by Hunter and Hash (1949).

LITERATURE REVIEW

Composition of the pegmatities over this entire region is remarkably uniform according to Parker (1949) with the typical rock consisting essentially of plagioclase, perthitic microcline, quartz and muscovite. The pegmatites and granites are weathered to depths up to one hundred feet

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depending on the topographic location. Deeply weathered deposits are located on river terraces; it is in these deposits that transitions from completely altered feldspar grading into parent material are exposed by mining operations.

According to Parker (1946) the most extensive period of kaolinization in the Spruce Pine district of western North Carolina occurred during the period in which the terraces were formed. The altitude of these terraces led Bridges (1949) to suggest that they could be considered as intermontane extensions of the Harrisburg peneplane of early Tertiary age. Bridges states that rocks in the Blue Ridge region were weathered deeply during the interval beginning with the uplift of the Schooley or Kittatinny peneplane and throughout the period in which the Harrisburg peneplane was being formed. Hunter (1940) and others observed that the depth of alteration coincides with the lowest level at which water freely circulates and the clay deposits occupy such a position relative to the river levels.

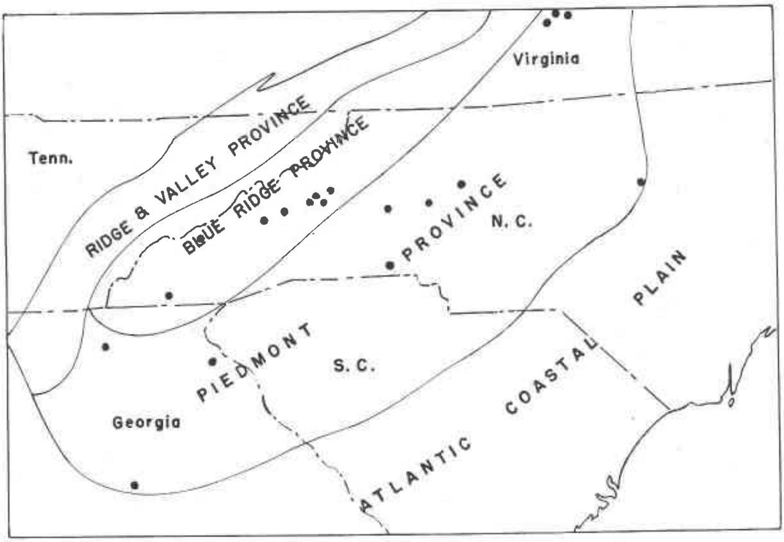

FIG. 1. Areal map showing locations of deposits sampled.

Ross and Kerr (1930, 1934), in their classic contributions concerning minerals of the kaolin group, state that halloysite very commonly is associated with kaolinite in some deposits, especially residual deposits derived from pegmatites. They suggest that halloysite occurring under such conditions might represent a stage in the eventual crystallization of kaolinite. Hunter and Hash (1949), conversely, suggest that in the halloysite deposits of western North Carolina alternate wetting and drying, with the possibility of aeration, might have been influential in the conversion of kaolinite to halloysite. Hunter (1940) previously stated that optimum conditions for kaolinization obtain where the topographic features encourage drainage and where the feldspathic rock is covered by dense vegetation. Cady (1950) reports on two deposits in the Piedmont province of North Carolina; he believes halloysite is formed in regions where weathering is slow and free movement of air and water is restricted.

Early workers on these deposits (Ries, 1911; Lindgren, 1915; and Bayley, 1921) all attributed the formation of these clays to weathering. Regarding the alteration of the mica, Bayley (1925) states that near the surface the mica is altered and loses its characteristic features; he designated the decomposition product "hydromica." Selle (1876), Hickling (1908) and Galpin (1912) advocated the origin of kaolinite through the formation of mica as an intermediate product. Galpin states that it is probable that secondary muscovite, representing the first stage of the weathering of feldspars, is converted directly into kaolinite. Regarding the North Carolina deposits, Ross and Kerr (1930) call the intermediate product a muscovite-like kaolin mineral. They state that crystals form vermicular aggregates like those of kaolinite in the same deposit and that many of these groups are partially altered to kaolinite. Denison, Fry and Dile (1929) studied the alteration of mica in soil profiles and concluded from chemical and optical evidence that the muscovite alters to kaolinite.

In 1943, the relation between hydrated halloysite (endellite) and its dehydrated product (halloysite) was elucidated by Alexander, *et al.* The tubular morphology of halloysite was determined by Bates, *et al.* (1950).

METHODS OF INVESTIGATION

Analyses of dry clay samples collected in 1950 disclosed the need for keeping the clays in their original hydrated condition and for the development of quantitative techniques. Moist samples were collected in 1951 with Professor T. F. Bates of The Pennsylvania State University.

A differential thermal analysis technique was developed which identified the amount of hydrated halloysite and kaolinite in a sample (Sand and Bates, 1953). Techniques used for quantitative analysis of dry samples containing halloysite and kaolinite are described by Sand and Ormsby (1953).

To determine quantitatively the mineral content of the residual clays, two corroborative methods were used; namely, point counts of thin-sections and analyses of size fractions by *x*-ray diffraction and glycol-differential thermal analysis. The point counter method, as modified by Chayes (1949) was applicable since, in most of the samples studied, the kaolinite occurred as vermicular aggregates easily distinguished from halloysite. Where the mica was in an intermediate stage of decomposition, it was difficult at times to differentiate between the interleaved kaolinite and parent mica. Size fractionation was employed to make mineral separations on the basis of particle size. Quartz and feldspar generally concentrated in the coarsest fractions, mica and kaolinite in intermediate fractions and hydrated halloysite in the finest fractions. The samples were mulled with a rubber pestle, the suspension brought to $pH=10$ by addition of ammonium hydroxide and blended for five minutes. This treatment did not disaggregate the hydrated halloysite completely, but did prevent the disaggregation of the relatively coarse "books" of kaolinite and mica. Wet-sieve separations were made through 60 and 200 mesh screens and gravity separations were made through the 8 micron size fraction. For separating finer fractions, centrifuge methods were used. Size frequency distributions were obtained simultaneously. Each size fraction was analyzed by *x*-ray diffraction, differential thermal and petrographic techniques to obtain quantitative mineral constitution. The Norelco *x*-ray spectrometer was used; peak intensities were used to estimate percentages of minerals present. The moist sample first was analyzed and then dried at 100° C. to resolve the hydrated halloysite and mica basal reflections which mutually interfere.

In studying deposits of halloysite, it is advantageous to keep the samples moist for analysis. Allowing samples to become even partially dehydrated makes quantitative analysis difficult except for petrographic and electron microscope techniques. Estimates of halloysite content on the basis of electron micrographs is hazardous if the kaolinite is present as vermicular aggregates in a coarse size range. Special preparation, as by homogenization, offers some possible quantitative use for the electron microscope on such samples. In those samples where the vermicular habit of kaolinite is not well-developed, the thin-section method for quantitative determination of kaolinite versus halloysite is useless. Analysis of the moist sample by the *glycol-DTA* method is preferred in such samples. If either the kaolinite or hydrated halloysite content is low, size fractionations should be made and the fractions analyzed.

ANALYSIS OF SAMPLES

Virginia Piedmont Province

Samples from four deposits in this region were examined. They were found to be principally kaolinite with some hydrated halloysite present in minor amounts. In each of the deposits, the feldspar was observed altered in part to secondary mica from which vermicular kaolinite was being derived. Samples from three of the deposits were studied in thin-section. An analysis of size fractions was made of a sample from the Nat Kidd Prospect in Roseland. This analysis is given in Table 1 to illustrate the fractions separated and analyzed. Complete analyses of other samples so analyzed are given elsewhere (Sand, 1952).

Table 1. Mineralogical Analysis of Size Fractions, Nat Kidd Prospect, Roseland, Virginia

Size fraction (microns)	Quartz	Mica	Feldspar	Hydrated halloysite	Kaolinite	Size analysis
+250	tr.	0.2%	tr.		tr.	0.2%
74–250	0.1%	0.3			0.2%	0.6
31–74		6.1			5.9	12.0
16–31		13.2			13.4	26.6
8–16		8.6			19.0	27.6
4–8		3.1			12.3	15.4
2–4		1.3			7.2	8.5
1–2		tr.		0.1%	0.7	0.8
0.5–1		0.1		0.8	2.1	3.0
–0.5		0.2		1.4	3.7	5.3
Bulk Composition	0.1	33.1		2.3	64.5	100.0

The bulk composition of this sample correlated with the thin-section analysis of a sample from the American Rutile Mine in Roseland which contained 70% kaolinite, 29.2% mica and 0.8% quartz. This sample had dehydrated and although the amount of halloysite was not determined, only a few halloysite particles were present among kaolinite "books" and plates in the electron micrograph.

Samples taken from the Calco Ilmenite Mine at Piney River, and the Morefield Mine at Amelia Court House, were found to be similar in mineral constitution. The secondary mica in all these deposits formed in a fine-sized vermicular habit by alteration of the feldspar. The mica in turn was weathered to vermicular aggregates (about 0.02 mm. size) of kaolinite. The vermicular habit was not as pronounced as in deposits further south.

North Carolina Piedmont Province

Qualitative examination of samples from gneissoid rocks in this region showed a predominance of kaolinite over halloysite. In these samples no vermicular habit was observed, nor was it evident that mica was an intermediate product. The small amount of halloysite present in these samples occurred as well-developed tubes up to ten microns in length.

Two moist samples from the Patterson Mica Mine were analyzed in detail. One sample consisted of a granitic material obtained thirty feet below the original surface. The kaolinite appeared in the coarsest size fractions due to interleaving with the parent mica. The bulk composition, as determined by analysis of the size fractions, is listed in Table 5. The other sample was a decomposed feldspar crystal obtained from pegmatitic material located 50 feet below the original surface. Its bulk composition is also listed in Table 5. The large variation that can obtain between clay samples in one deposit is demonstrated by these two analyses.

North Carolina Blue Ridge Province (Northern Region)

Eight deposits were studied in this region usually designated as the Spruce Pine pegmatite district. Of these, the Gusher Knob and Micaville deposits are in granitic bodies cut by pegmatite veins; the others are in pegmatites.

Hydrated halloysite predominated over kaolinite in all of these deposits. In individual samples the hydrated halloysite to kaolinite ratio varied from 30:1 to 1:1 with an average for the region of about 10:1. The feldspars, whether sodic plagioclase or microcline, weather to hydrated halloysite with plagioclase the first to decompose. This can be observed in the hand specimen of a partially altered perthite where the potash feldspar stands out in relief. Large, only slightly altered microcline crystals were found next to completely altered plagioclase. However, in this region it was found that all feldspars alter to hydrated halloysite even though the rate of weathering differs. In only one sample, from the Woody Deposit, was the potash feldspar observed altered in small part to secondary mica. The primary mica was altered to vermicular kaolinite in all samples studied.

In each of the two granitic bodies, a study was made on vertical profiles of the degradation of muscovite. In the Micaville deposit, the mica content in the finer fractions decreases upward in the soil profile. To illustrate this, the mica and feldspar contents (per cent of total composition) are excerpted from the analyses of the size fractions of sample S-21 (top), S-23 (middle) and S-27 (bottom) as shown in Table 2.

The disappearance of the muscovite in the finer fractions also was found to obtain in four vertical sections taken from a sample grid in the Gusher Knob deposit. The vertical profiles each contain three samples separated by three foot intervals. To illustrate, Table 3 lists the mica

TABLE 2. MICA AND FELDSPAR CONTENT IN A VERTICAL PROFILE, MICAVILLE DEPOSIT

Size fraction (microns)	Mica			Feldspar		
	Top	Middle	Bottom	Top	Middle	Bottom
+250	6.4%	6.2%	0.4%	none	none	1.1%
74–250	5.1	23.4	0.7		0.5%	3.7
31–74	1.1	3.6	10.6		0.2	15.2
16–31	1.4	3.1	2.6			2.6
8–16	0.5	2.2	2.9			0.8
4–8	0.2	0.5	1.1			0.2
2–4	0.1	0.3	0.5			
1–2		0.2	0.2			
0.5–1		0.1	0.1			
–0.5		tr.	0.3			

content (per cent of total composition) in one typical profile in the finest fraction in which mica occurs.

Because of the wide variability in total content and size distribution of muscovite between samples, inverse correlation of total mica content with total kaolinite content does not always obtain. However, the mica in the finest fractions, which is the first to alter completely, was observed to disappear upward in the profile. The grid sampling taken in the Gusher Knob Deposit was in a weathered, fairly uniform granitic body at the east end of the East pit. The samples were taken three feet apart on the vertical and ten feet apart along the horizontal. Samples 52 and 62 were the top samples in the central vertical profiles. Bulk mineral compositions, as obtained from analyses of the size fractions, are given in Table 4 to show the large sample variation.

North Carolina Blue Ridge Province (Central Region)

In Buncombe County, completely altered pegmatites, selected for detailed study, showed a general decrease in the content of hydrated

TABLE 3. MICA CONTENT IN THE FINEST FRACTION IN WHICH MICA OCCURS, GUSHER KNOB DEPOSIT

Sample No.	Size Fraction	Mica Content
50 top	8–16 microns	2.0%
47 middle	8–16 microns	7.5%
42 bottom	8–16 microns	10.0%

TABLE 4. BULK MINERAL COMPOSITION BY ANALYSIS OF SIZE FRACTIONS, GUSHER KNOB DEPOSIT

Sample No.	Quartz	Mica	Feldspar	Hydrated halloysite	Kaolinite
West Wall of Grid					
42-bottom	0.8%	17.1	0.7	66.2	15.1
47-middle	7.7	32.6	none	55.4	4.3
50-top	10.3	18.6	none	67.4	4.1
44-bottom	23.3	24.3	3.9	41.3	7.0
45-middle	16.6	14.8	0.9	55.7	12.1
52-top	15.8	8.4	none	62.5	12.3
South Wall of Grid					
54-bottom	45.0	15.2	none	29.0	11.8
59-middle	37.5	16.4	none	21.5	24.7
62-top	30.2	10.8	none	42.6	16.5
56-bottom	32.3	12.1	none	42.5	13.2
57-middle	30.8	17.7	none	31.7	19.8
64-top	38.5	11.2	none	42.3	8.0

halloysite and a corresponding increase in the amount of kaolinite over those from the Spruce Pine District. Most of the feldspar crystals, whether plagioclase or microcline, alter to hydrated halloysite. However, some of the microcline in each deposit alters to secondary mica which weathers to kaolinite. Bulk mineral compositions from the Arrowhead and Alexander deposits are given in Table 5.

North Carolina Blue Ridge Province (Southern Region)

An increase in the amount of kaolinite derived from feldspar was noted in this region. In one sample from near Hayesville, the hydrated halloysite to kaolinite ratio was 1:23. This was the only sample from this region that was so high in kaolinite content. In all other deposits in this region, including Macon, Swain, Haywood and Clay counties, the feldspar alters in part to secondary mica and the remainder to hydrated halloysite. In a perthite crystal the plagioclase alters principally to hydrated halloysite and the microcline principally to mica and eventually to kaolinite.

The Georgia Piedmont

The kaolin residual clays in this province showed a predominance of kaolinite over hydrated halloysite. The ratio of hydrated halloysite to

TABLE 5. BULK MINERAL COMPOSITIONS

Deposit	Analysis	Parent rock	Quartz	Mica	Feldspar	Hydrated halloysite	Kaolinite
Va. Piedmont							
Nat Kidd	S F.	syenite	0.1	33.1	tr.	2.3	64.5
N.C. Piedmont							
Patterson	S.F.	granite	5.0	35.1	none	0.5	59.5
Patterson	S.F.	pegmatite	0.6	11.0	none	27.1	61.3
N.C. Blue Ridge (Northern)							
Gusher Knob							
Average, 12 samples	S.F.	granite	24.1	16.4	0.5	46.6	12.4
School Hill	S.F.	granite	58.2	1.9	none	35.3	4.9
Fluken Ridge	T.S.	pegmatite	22.4	0.3	0.8	73.7	2.8
Fluken Ridge	T.S.	microcline	0.4	1.3	0.3	90.8	7.2
Fluken Ridge	S.F.	microcline	0.7	7.0	4.7	74.4	13.1
Fluken Ridge	S.F.	perthite	tr.	6.4	39.7	52.2	1.7
Fluken Ridge	S.F.	perthite	4.6	2.1	75.6	16.3	1.4
17-Acre Pros.	S.F.	pegmatite	5.5	16.8	none	64.5	13.2
17-Acre Pros.	S.F.	pegmatite	8.8	18.2	none	61.8	11.1
17-Acre Pros.	T.S.	pegmatite	3.0	17.1	none	66.8	13.1
17-Acre Pros.	S.F.	pegmatite	3.4	8.3	none	75.5	12.9
17-Acre Pros.	T.S.	pegmatite	1.8	3.8	none	85.6	8.8
17-Acre Pros.	T.S.	pegmatite	none	6.3	none	84.9	8.8
Carter Ridge	T.S.	pegmatite	none	2.9	none	84.3	12.8
Micaville	S.F.	granite	4.4	14.8	none	78.0	2.7
Micaville	S.F.	granite	29.9	39.7	0.7	25.1	4.7
Micaville	S.F.	granite	5.7	19.4	21.6	43.1	10.2
N.C. Blue Ridge (Central)							
Arrowood	T.S.	pegmatite	none	0.6	13.9	75.1	10.4
Arrowood	S.F.	pegmatite	1.2	3.9	7.5	69.2	21.2
Alexander	T.S.	perthite	none	1.6	0.2	87.5	10.7
Alexander	T.S.	perthite	none	2.7	none	86.8	10.5
Alexander	T.S.	microcline	none	0.4	0.2	91.8	7.6
Alexander	S.F.	perthite	0.9	7.3	0.6	75.0	16.1
Alexander	S.F.	perthite	3.3	8.8	none	70.0	17.9
Alexander	T.S.	perthite	0.9	6.3	0.8	74.3	16.7
Alexander	S.F.	perthite	0.6	20.5	none	17.0	7.8
Alexander	T.S.	perthite	none	4.6	none	88.9	5.6
Alexander	S.F.	perthite	none	13.4	none	67.6	18.9
N.C. Blue Ridge (Southern)							
Whitehall	T.S.	pegmatite	none	20.0	0.4	71.2	8.4
Bethel	T.S.	pegmatite	none	1.6	none	76.5	21.8
Cox	S.F.	granite	22.2	32.9	4.8	1.7	39.4
Georgia Piedmont							
Funkhauser	S.F.	pegmatite	13.9	41.2	0.3	1.6	42.9
Funkhauser	S.F.	pegmatite	38.2	12.6	none	3.0	46.5
Funkhauser	S.F.	pegmatite	3.8	5.0	23.0	17.4	51.0

S.F.—Analysis of size fractions.
T.S.—Point-count of thin-sections.

kaolinite was 1:27 in one sample and averaged about 1:6 in those samples analyzed. Most of the feldspar alters to vermicular kaolinite through the intermediate mica stage and the remainder alters to hydrated halloysite. The primary mica alters to vermicular kaolinite as was the case in all the deposits studied. Bulk analyses of samples from the Funkhauser Mine are listed in Table 5.

DISCUSSION

The sequence of alteration of minerals follows that given by Goldich (1938). The plagioclase alters first, followed by potash feldspar and mica. Quartz was not altered noticeably. In thin section the feldspars are observed altered to halloysite. Remnants of the same crystal of feldspar are present in a matrix of halloysite. Remnants of feldspars, partially altered to "books" of secondary mica and kaolinite likewise were observed. The absence of fine-grained kaolinite in most of the samples was evidenced by the good agreement obtained in the same sample between the point-count method and the analysis of size fractions. In general, the "books" of mica derived from weathering of feldspar were smaller than primary mica and did not show as high a retardation. The "books" of secondary mica were not arranged parallel to each other. This resulted in a random arrangement of vermicular kaolinite aggregates as contrasted to the oriented aggregates derived from a larger "book" of primary muscovite. When the latter is kaolinized, the "books" show a parting perpendicular to the basal cleavage at 0.02 to 0.04 mm. intervals. The kaolinite was found to concentrate in this size fraction. Books of mica, which were washed thoroughly and kept in a moist condition, were found to be muscovite and kaolinite. In no such sample, even though the mica was embedded in a matrix of hydrated halloysite, was the halloysite found interleaved with the mica. The kaolinite is pseudomorphous after the mica, and the basal planes of the kaolinite orient, in general, parallel to those of the mica. Deviations occur as a result of parting and a curving of the vermicular kaolinite aggregates as the mica is altered. The interleaving of the mica and kaolinite was on a gross scale; no mixed-layer effects were observed.

Residual kaolin clays derived from feldspathic rocks in the Southern Appalachian show a general increase in the content of hydrated halloysite southward from Virginia into North Carolina where a maximum of hydrated halloysite is noted in the Spruce Pine district. Southward from this district to the southern end of the Appalachian chain, there is a general decrease in the amount of hydrated halloysite until at the southernmost deposit studied, kaolinite predominates. Although these are the general trends, locally there are exceptions, since primary mica content, topographic expression, drainage and vegetation cover are all influencing factors in addition to that of overall climate.

Hydrated halloysite, as observed, had formed by the weathering of feldspars. No transitions between hydrated halloysite and kaolinite were evident. Effective leaching from the feldspar of all of its bases probably destroys its structure and the colloidal silica and alumina are arranged into the random structure of hydrated halloysite. It is significant that hydrated halloysite, not its dehydrated product, always was present except where samples were obtained from dried exposures. This indicates that environmental conditions conducive to the formation of hydrated halloysite have not changed appreciably since Tertiary time. The stability of hydrated halloysite up to 100° C. was determined by Sand (1952). The boundary curve delineating the stable field for hydrated halloysite follows fairly closely the vapor pressure-temperature curve of water. Although hydrated halloysite does dehydrate rather easily as reported by MacEwan (1946), Brindley and Goodyear (1948) and others, it can exist to relatively high temperatures as shown by Roy and Osborn (1954). Hydrated halloysite was not observed to coexist at equilibrium in various proportions with halloysite at various humidities as has been reported. Theoretically, only on the univariant boundary curve can they coexist as stable phases, presuming a starting material of pure hydrated halloysite. If halloysite is present in a sample of hydrated halloysite due to partial dehydration, then halloysite will coexist with the hydrated form since the reaction is irreversible. Brindley also proposes the phenomenon of entrapped pockets of hydrated halloysite present in halloysite.

No matter what the intensity of leaching, even where the feldspars alter completely to hydrated halloysite, the primary muscovite mica always alters to kaolinite. Thus, in a completely weathered perthite crystal, patches of vermicular kaolinite derived from the disseminated mica "books" occur in a matrix of hydrated halloysite. This is an apparent anomaly which can be explained only on the basis of structural control. Once the feldspar alters to secondary mica, the compositional and structural control, evidently imposed by the mica, causes vermicular kaolinite to be the next weathering product. A completely leached and hydrated muscovite has the same ratio of alumina, silica and water as kaolinite, but the mechanism of transfer of silicon and aluminum from a 2:1 layer structure to a 1:1 type is unknown. The vermicular kaolinite in these deposits usually is not present in size fractions below two microns; a relatively pure hydrated halloysite can be obtained by fractionation.

The nature of the clay end product resulting from the weathering of feldspathic rocks in this region is believed to be a function of mineral composition, rock texture, degree of fracturing, topographic expression, vegetation cover and climate. Where conditions favor the formation of hydrated halloysite, a feldspathic rock high in mica content yields a kaolin clay correspondingly high in kaolinite. Conversely, a rock low in mica yields a clay high in content of hydrated halloysite. For this reason, some samples are lower in hydrated halloysite content in the Spruce Pine district as compared to some pegmatitic samples from the Piedmont. Rock texture and degree of fracturing determine the drainage pattern. Where vegetation cover was sparse a tendency toward formation of secondary mica was noted. River terraces apparently are favorable locations for the formation of extensive deposits of hydrated halloysite. Good drainage effects a thorough leaching of potash and inhibits the formation of secondary mica.

This work concurs with the early views of Selle and others that secondary mica is an essential intermediate product in the formation of residual kaolinite from feldspar.

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