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Occurrence of dickite in Jamaica—ordered and disordered varieties

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Abstract

The geology of the area near Job's Hill, St. Mary, Jamaica, is reviewed with reference to the occurrence of dickite, which appears to have been formed by hot ascending waters or emanations from igneous bodies along fractures and through brecciated zones in at least three stages. Angular fragments of the initially-formed dickite, mainly pink, are embedded in a matrix of subsequently-formed dickite, mainly green; narrow veins and coatings of dickite, often white, also occur.

X-ray diffraction patterns of dickites show a range of order–disorder; the pink angular fragments are often highly disordered, the green matrix material disordered to various extents, and the white coatings may be well-ordered or variously disordered. Disorder weakens or eliminates diffractions with the index $k \neq 3n$, and the most highly disordered dickites are difficult to distinguish from highly disordered kaolinites. The structural relations of dickite and kaolinite and the probable nature of the disorders are reviewed.

DTA patterns of the Jamaican and other dickites exhibit endotherms ranging from broad almost-double peaks extending from 500–700°C to sharp peaks near 700°C. There is a tendency to pass from broad to sharp endotherms with increasing structural order, but exceptions are found, so that no firm rule can be claimed. The highly anomalous result given by the well-ordered Keokuk geode kaolinite, namely a DTA pattern resembling those of disordered dickites, is confirmed, but no obvious explanation is forthcoming.

Introduction

Material resembling dickite at Job's Hill, St. Mary, Jamaica was described by Davis *et al.* (1970) as "this remarkable and possibly unique deposit," but they did not explain why it might possibly be unique. X-ray diffraction patterns indicated that the material belonged to the kaolinite group, but the high degree of sample orientation shown by the published patterns precluded any detailed consideration of the structure. DTA curves showed endotherms of the type commonly found with dickites, and infrared spectroscopic data also pointed to dickite rather than to kaolinite. Their final conclusion was that the material was "essentially dickite probably with associated nacrite and kaolinite."

A sample of the Job's Hill material given to one of us (G.W.B.) by Mr. D. A. Holdridge in 1970 provided an X-ray pattern which suggested a type of crystallization intermediate between those of dickite and kaolinite. Subsequent field observations by one of us (A.R.D.P.) led to the discovery of other dickites with various degrees of structural order–disorder. While disordered forms of kaolinite have been described many times, few accounts of disordered dickites have appeared. Hayes (1967) and Schroeder and Hayes (1968) described dickites and kaolinites in limestones of southeastern Kansas and found that structural disorders occur in the dickites but "the range of variation is not as great as for the associated kaolinites." The Kansas dickites were said to be unusual in that they formed "in rocks that were neither deeply buried nor extensively altered hydrothermally."

It has been known for at least 20 years that the DTA curves of dickites are often strangely different

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from those of kaolinites, particularly as regards the form of the endothermic peak which corresponds to the dehydroxylation reaction; this peak may be broad, almost double, and extend from about 500–700°C, or a sharp peak near 700°C, or some intermediate form. The range of order–disorder found in the Jamaican dickites permits further consideration of this phenomenon.

Geology and occurrence of Jamaican dickite

Three mappable units are recognized in the immediate vicinity of Job's Hill (Fig. 1).

The Border Volcanic Formation of Upper Cretaceous age This comprises a complex sequence of massive to poorly-bedded andesitic breccias, conglomerates, agglomerates and tuffs, with intercalated andesitic flows and minor intrusives. These rocks are locally mineralized, and exposures of dickite are confined to two narrow, roughly parallel northeast–southwest trending zones of hydrothermally altered rock southeast of Job's Hill.

Granodiorite Towards the end of the Cretaceous period, widespread intrusions of granodiorite occurred (Chubb, 1962, pp. 20,21), accompanied by thermal metamorphism raising some of the rocks within narrow contact zones outside of the study area to hornblende- and pyroxene–hornfels facies (Reed, 1966). Away from these zones, plagioclase and ferromagnesian minerals are only partly altered to typical low temperature minerals such as chlorite, epidote and calcite. Subsequent uplift and erosion unroofed these intrusions prior to deposition of the earliest Tertiary sediments.

The Wagwater Formation of Lower to Middle Eocene age This formation, which unconformably overlies the Cretaceous succession in some places and is faulted against it in others, consists of a sequence of well-bedded conglomerates, sandstones, and siltstones varying in color from purplish to reddish brown. Although this formation is largely unfossiliferous, boulders and pebbles of Cretaceous limestone and granodiorite serve to define its stratigraphic position.

Northerly dickite-bearing zone

Trenching operations carried out by the Geological Survey Department in 1963 to determine the extent of the northerly dickite-bearing zone at G.R. E6000 N4645 (the "northern vein" of Davis *et al.*, 1970) revealed a maximum width of 21 m and an outcrop length of about 180 m. The dickite in this zone ranges from blocks of an indurated breccia containing cream to pinkish and purplish fragments composed largely of dickite with subordinate anatase/leucoxene set in a matrix of essentially green dickite, to discrete veins and surface coatings of white, cream, and translucent dickite. Polished and striated surfaces on some of the vein dickite are indicative of postdepositional movement.

The most northeasterly exposure is best described as a talus heap of massive blocks and smaller fragments of indurated dickite and purplish cataclasite, currently used as road metal. The extension of the zone can be traced in a southwesterly direction towards the top of a hill, where a large outcrop protrudes from the soil. On the northwestern side of the dickite-bearing zone is an area of greenish-grey to reddish-brown weathered malachite-bearing volcaniclastic rocks assigned here to the Border Volcanic Formation. To the immediate southwest is an area of copper mineralization which was drilled in 1956 by Burrex Mines Ltd.; the corehole locations are shown in Figure 1. According to A. J. Reed (unpublished report, February 1966) "light grey kaolin-rock" and a "small vein of kaolin-rock" were encountered in B.H. 3 and 4, respectively within a sequence of red andesitic agglomerate and fine-grained, purplish-grey tuff, but this kaolin rock is no longer available for examination.

It appears from all the available data that the dickites of the northerly zone were formed by hot ascending waters of uncertain origin and/or direct emanations from intrusive igneous bodies in at least three stages. The first is represented by brecciation of the country rock followed by incomplete dickitization of the broken fragments. Subsequent movement followed by dickitization of all open spaces constitutes the second stage. These forms of dickite are found to be poorly ordered. The third stage led to deposition of narrow veins and surface coatings of mainly well-ordered dickite.

Southerly dickite-bearing zone

Whilst the northerly zone has been studied previously, little attention has been paid to the smaller southerly zone located at G.R. E6005 N4630, where a 9 m wide shear zone dips steeply towards the northwest. Its lateral extent is approximately 60 m.

The character of the rock within this zone ranges from a hard grey feldspathic tuff with disseminated pyrite to a soft laminated almost schistose rock containing thin alternating laminae of white powdery well-ordered dickite and red earthy hematite; surface

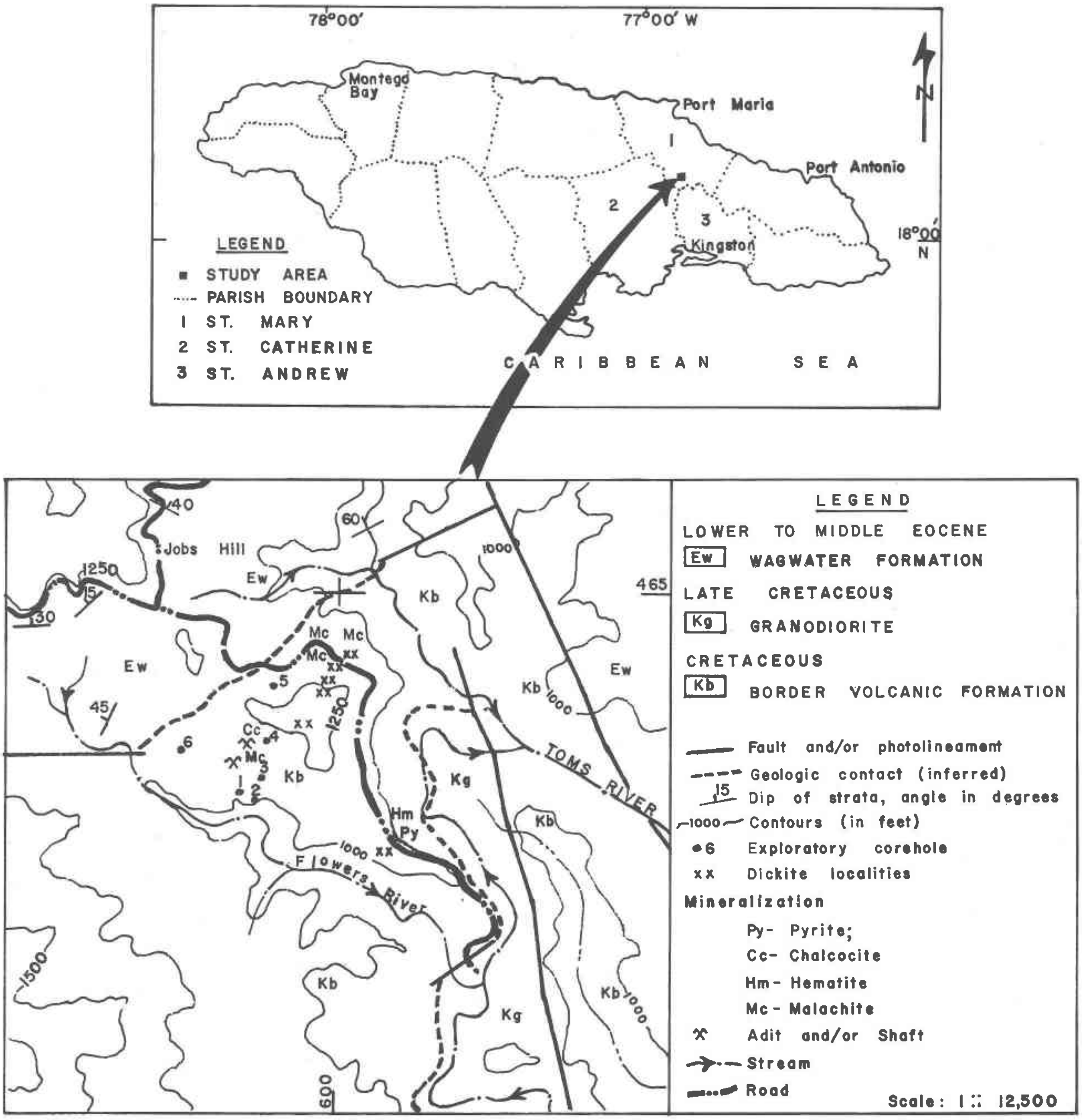


Fig. 1. Index map of Jamaica showing projection of dickite localities and simplified geological setting. (Modified after the unpublished work of Burrex Mines Ltd. and A. J. Reed.)

coatings of metallic hematite are a prominent feature of this zone.

Other occurrences

The only other references known to us regarding the possible occurrence of dickite in Jamaica is that of A. J. Reed of the Geological Survey Department, who mentions in an unpublished account on the Richmond quadrangle written in 1966 the presence of a light-grey indurated clay similar to that of Job's Hill within the plane of the Wagwater fault where it crosses Tom's River east of the study area.

X-ray diffraction data

Figure 2 shows representative X-ray diffractometer patterns (taken with Ni-filtered Cu$K\alpha$ radiation at

1°[2θ]/min) of samples from various localities near Job's Hill. In preparing the samples, pestle and mortar grinding was avoided. Powdery materials were picked from associated material with a needle point or razor blade, and hard materials were scraped with a razor blade. Samples were sieved, if necessary, through a 120 mesh screen and were mounted with a minimum of preferred orientation in shallow cavities about 1 × 3 cm^2 in glass slides. This relatively coarse powder was preferred to finer material disordered by grinding.

Figure 2A is a pattern of a structurally well-ordered dickite taken from a white rim on a massive sample from the northerly vein at Job's Hill. Similar patterns are obtained from white powdery samples taken from crumbly material collected from the southerly vein. The well-ordered samples show the full diffraction pattern expected from the crystal structure of dickite; the spacings and intensities agree well with the data listed by Bailey (1963) and indicate relatively little structural disorder in these materials.

Figures 2B, C, D show dickite patterns with increasing structural disorder. From all the materials examined, it seems that most of them, particularly the massive materials, contain appreciable or even considerable structural disorder. For comparison, Figure 2E shows the pattern of a highly-disordered kaolinite from Pugu, Tanganyika, previously studied by Robertson *et al.* (1954). The similarity of Figures 2D and E illustrates the difficulty of distinguishing highly-

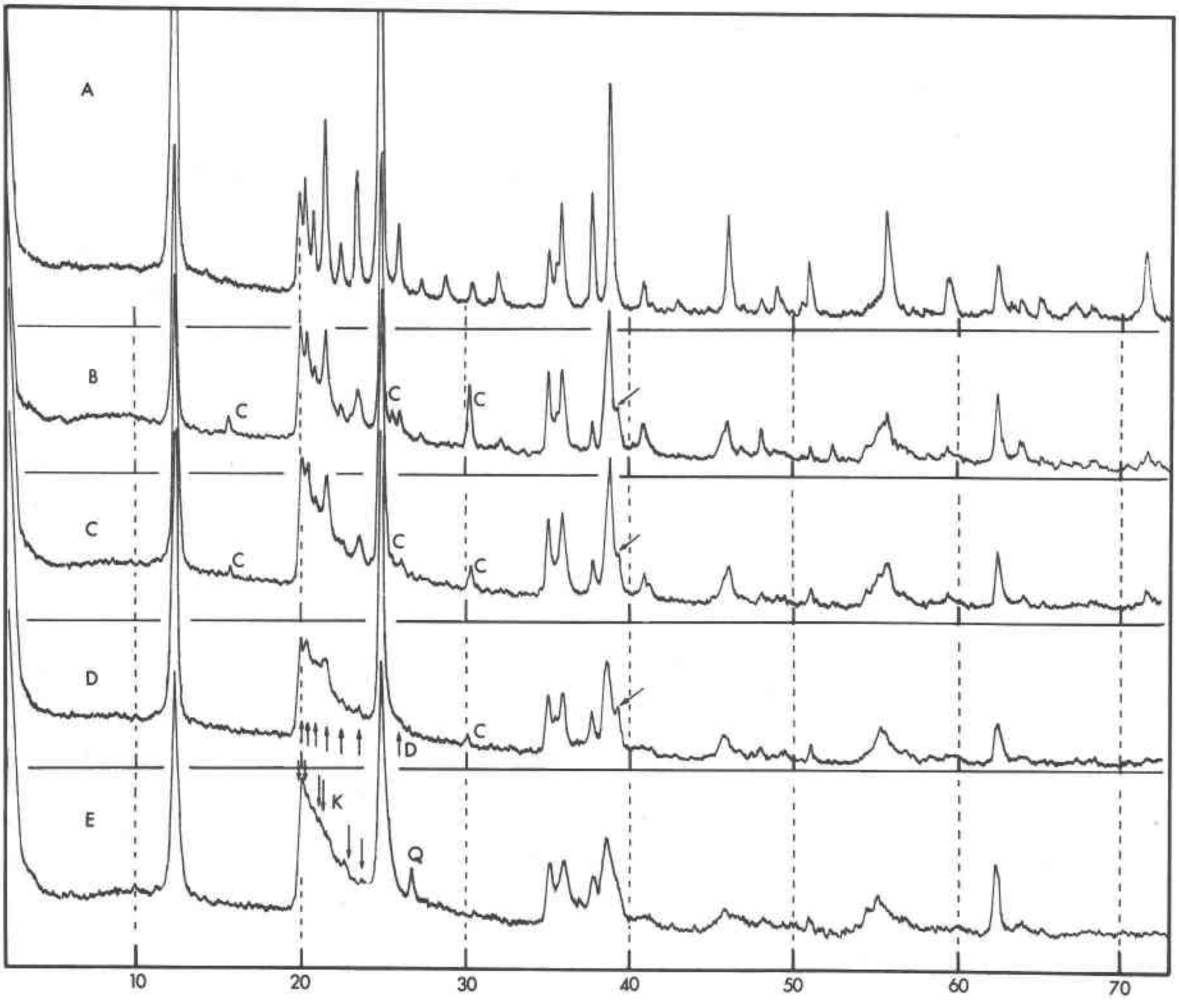


Fig. 2. X-ray powder diffraction patterns *versus* 2θ, degrees; Cu$K\alpha$ radiation. A. Dickite, Jamaica, sample #7, white surface coating, DTA curve 9. B. Dickite, Jamaica, sample #1, pale green material. DTA curve 7. C. Dickite, Jamaica, sample #3, white surface layer. DTA curve 3. D. Dickite, Jamaica, sample #5, pink color. DTA curve 8. E. Kaolinite, Pugu, Tanganyika (see Robertson *et al.*, 1954). Arrows marked D and K indicate positions of dickite and kaolinite reflections. C and Q indicate crandallite and quartz reflections. Arrows at $2\theta \simeq 39°$ on patterns B, C, D, indicate possible kaolinite-type diffraction (see text).

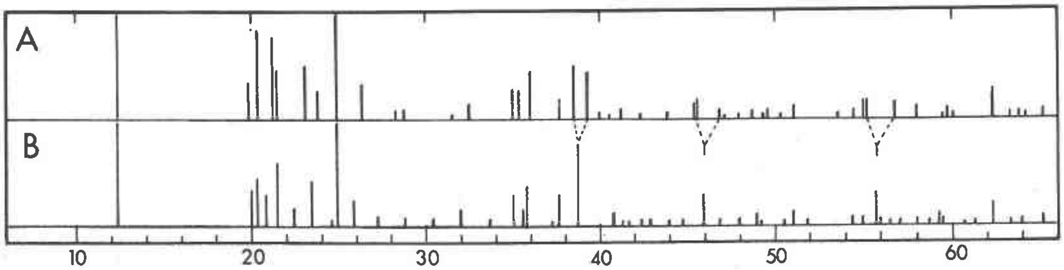


Fig. 3. Diagrammatic X-ray diffraction patterns *versus* 2θ, degrees; Cu$K\alpha$ radiation. A. Kaolinite, data for sample from Keokuk, Iowa. B. Dickite, observed and calculated data of Bailey (1963). Dotted lines show examples of reflections from monoclinic dickite split into pairs of reflections from triclinic kaolinite due to an α angle not equal to 90°.

disordered kaolinites and dickites; materials may well exist where a distinction is scarcely possible.

Identifying characteristics of dickite and kaolinite

The line diagrams in Figure 3 compare the powder-diffraction patterns of well-ordered kaolinite and dickite for Cu$K\alpha$ radiation. The dickite data are based on the observed and calculated values given by Bailey (1963, p. 1202). The kaolinite data are experimental data for the exceptionally well-ordered kaolinite from Keokuk, Iowa. These patterns are sufficiently different that well-ordered minerals of either kind are easily recognizable.

In dickites, as in kaolinites, structural disorder has the effect principally of weakening and broadening reflections with the index $k \neq 3n$. Between 20° and 27° (2θ), reflections of the type $11l$ and $02l$ occur with $l = 0,1,2$ for dickite and $l = 0,1$ for kaolinite which generate obviously different patterns. With increasing structural disorder, these reflections rapidly become weak and are replaced by a band of scattering with weak modulations. Between 27° and 34° (2θ) a sequence of almost equally-spaced weak to medium intensity reflections occur from dickite which are highly characteristic; these are $11l$, $02l$ reflections with $l = 3,4,5$ and they disappear even more rapidly with increasing disorder than those with $l = 0,1,2$. Between 34° and 40° (2θ), two groups of strong reflections with $k = 0$ and 3 commonly appear as two doublets from dickite and two triplets from kaolinite, but these reflections also become less clearly diagnostic as disorder increases.

These remarks are illustrated by the patterns in Figure 2. In Figure 2B the highly characteristic reflections between 27° and 34° (2θ) are already reduced to about half normal intensity; in Figure 2C they have nearly disappeared, and in Figure 2D they are absent. The reflections between 20° and 27° (2θ) are still just visible in Figure 2D and correspond to dickite as shown by the arrows, whereas in Figure 2E similarly weak reflections correspond to kaolinite, as shown by the arrows. The stronger reflections with $k = 0$ and 3 in the range 34° to 40° (2θ) are discussed later. The reflections marked C and Q in Figure 2 arise from impurities of crandallite, a calcium aluminum phosphate hydrate, and quartz.

Structural relations between kaolinite and dickite

Kaolinite and dickite have been regarded for many years as distinctly different crystallizations, but a closer relation between them was shown by Bailey (1963), when he referred the dickite structure to a monoclinic cell with $\alpha = 90.0°$ and $\beta = 103° 35'$; these angles are almost identical with those for triclinic kaolinite with $\alpha = 91.6°$ and $\beta = 104.8°$. The a and b parameters of the two structures are essentially the same and c (dickite) is approximately $2c$ (kaolinite). The departure of α from 90° in kaolinite is cancelled in dickite, because the two-layer unit cell provides alternately positive and negative displacements between layers. The main difference between the two crystallizations lies in the distribution of the vacant octahedral sites which, following Bailey's notation, are wholly B or wholly C in kaolinite (these correspond to indistinguishable enantiomorphic structures) and alternately B and C in dickite.

Structural disorders in kaolinite have been discussed in relation to X-ray data particularly by Plançon and Tchoubar (1975, 1976) and by Plançon (1976), and their most recent conclusions, freely translated, are as follows: "The principal type of defect in the entire series of natural kaolinites is the displacement from one layer to another, or from one domain to another of the same layer, of the vacant octahedral sites. Stacking faults by translations of $\pm b/3$ arise as a secondary consequence of the displacement of vacant sites. A model involving faults due to rotations of layers is rejected."

In terms of Bailey's notation, the sequence of layers in dickite can be represented by ...BCBCBCB... and in kaolinite by ...BBBB... or ...CCCC... A "mistake" in the dickite sequence of vacant sites, such as ...BCBBBCB..., introduces a kaolinite component, and a "mistake" in the kaolinite sequence introduces a dickite component. The situation is less simple if A sites also become vacant. However, as the proportion of mistakes in either mineral increases, the end result will tend to be the same; patterns of the type shown in Figures 2D and 2E evidently approach this situation.

The angle α as a measure of "triclinicity"

Another aspect of the effect of "mistakes" concerns the angle α. In a sequence such as ...CBCBBBCBC... the dickite components ...CBCB... will be slightly offset by the triclinic angle α of the kaolinite block BBB. The dickite components may diffract independently or coherently, depending on the sharpness of their individual diffractions and the extent to which they are separated by the "foreign" block of kaolinite. Similarly, a dickite inclusion in a mainly kaolinite structure may diffract separately or as part of disordered kaolinite.

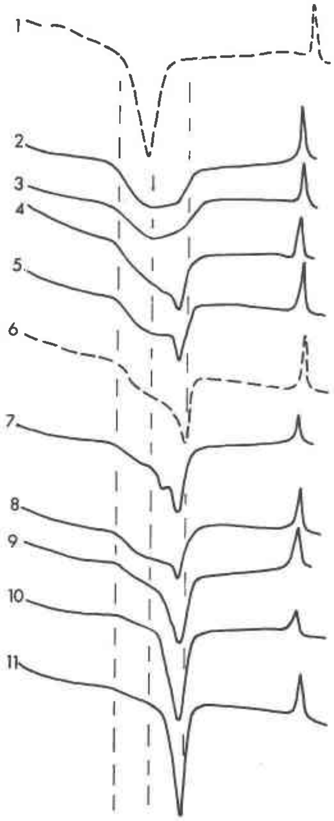
Plançon (1976, p. 49) suggests that a coefficient determining the monoclinic or triclinic character of the diffracting system may be obtained by considering the $1\bar{3}1$ and 131 diffractions from kaolinite; these reflections coalesce into a combined 131 for a single-layer monoclinic structure or 132 for the two-layer dickite structure. In Figures 2B, C, D an arrow points to a reflection apparently just emerging from the high-angle side of the 132 dickite reflection. This reflection cannot be indexed with the dickite lattice, but may represent the emergence of a $1\bar{3}1$ "kaolinite" (or triclinic) peak arising from an average value of α different from the monoclinic value of 90°. The 132 dickite reflection may move towards the 131 kaolinite reflection on the low-angle side. The relative positions of these reflections are seen in Figure 3. Slow recording of the diffraction patterns in the range 38°–40° (2θ) in order to measure the angular positions of the peaks accurately, however, has given only inconclusive results. If the relative positions of the 131 and $1\bar{3}1$ reflections of kaolinite and the 132 reflection of dickite depended only on the α angle, then the dickite reflection would be midway between the two kaolinite reflections. However, Figure 3 shows that the actual relationship is different, so that other variables also must be involved. The β angles of kaolinite (104.8°) and dickite (103.6°) are not identical, and possibly other variations such as the precise values of the layer spacings may be involved. Consequently there seems to be no simple way of estimating the proportion of misplaced layers in kaolinites or in dickites by this approach. Qualitatively, disordered kaolinite gives a diffraction pattern which approaches a pseudo-monoclinic arrangement, as Brindley and Robinson (1946) discussed much earlier.

Thermal analysis data

DTA curves of dickites differ remarkably from those of kaolinites in the temperature range 500–700°C where dehydroxylation occurs. Kaolinites, with one notable exception, show a single endotherm which is more or less sharp with a peak temperature around 550–600°; curve 1 in Figure 4 is typical. The positions and shape of the peak vary only slightly with structural order, and even DTA curves for halloysites do not differ radically from those for kaolinites; see for example the discussions given by Robertson *et al.* (1954). The notable exception among kaolinites is shown by curve 6 in Figure 4, given by the geode kaolinite from Keokuk, Iowa, which was discussed by Keller *et al.* (1966). The kaolinite curves in Figure 4 are shown as dashed lines to distinguish them readily from the dickite curves with full lines.

Dickites give variable endotherms ranging from broad peaks which extend from about 500°–700°C to sharp peaks near 700°C. To explore this range of thermal behavior more fully, a number of dickites have been examined from various sources as well as those from Jamaica. A representative collection of these curves is given in Figure 4, where they are arranged sequentially from the broadest to the sharpest endotherms observed; the samples used are indicated in the caption to Figure 4, where information relating to structural order is also given. The curves were obtained with a DuPont Thermal Analyser, model 900, with samples of 35–40 mg heated at 10°C/min in air. The temperature calibration is not exactly identical for all runs but is closely similar. The following discussion, however, is concerned with the general features of the diagrams rather than with exact temperatures.

Very broadly and with several exceptions it can be concluded that the sequence from curve 2 to curve 11 corresponds to increasing structural order. Curves 2, 3, 4 and 7 are from moderately-disordered samples giving X-ray patterns similar to Figures 2B and 2C, and curves 9, 10 and 11 from well-ordered samples giving X-ray patterns like Figure 2A. However, important exceptions must be noted. Curve 5 comes from a sample giving an ordered X-ray pattern identi-



DTA curve	*Mineral*	*Source and sample color*	*X-ray data*
1	Kaolinite	Florida, white	Well-ordered structure
2	Dickite	Jamaica, #4, white	Similar to Fig. 2B, crandallite prominent
3	Dickite	Jamaica, #3, white surface material	Fig. 2C, little crandallite
4	Dickite	Jamaica, #7, pale green	Similar to Fig. 2B, crandallite prominent
5	Dickite	Nowa Ruda, Poland, bluish-green	Similar to Fig. 2A, little or no impurity
6	Kaolinite	Keokuk, Iowa, white	Exceptionally well-ordered structure, no impurity
7	Dickite	Jamaica, #1, pale green	Fig. 2B, crandallite prominent
8	Dickite	Jamaica, #5, pink	Highly-disordered, Fig. 2D, crandallite trace
9	Dickite	Jamaica, #7, white surface material	Well-ordered structure, Fig. 2A, little or no impurity
10	Dickite	Red Mountain, Ouray, Colo., API #14	Well-ordered, similar to Fig. 2A, no impurity
11	Dickite	Schuylkill, Penna., white	Well-ordered, similar to Fig. 2A, no impurity

Fig. 4. Differential thermal analysis curves of two kaolinites (dashed curves) and nine dickites (full curves). Temperature scale approximate.

cal with 2A, and curve 8 from the sample giving the most highly disordered X-ray pattern, 2D.

Smithson and Brown (1957) studied dickite from Anglesey, Wales, where dickite was first discovered, and observed a DTA curve similar to curve 5. Their X-ray data suggest that the sample was structurally well-ordered. They were unable to find any appreciable kaolinite impurity and concluded that "the double endotherm must be tentatively attributed to dickite." Stoch (1964) studied several dickites, including one from Nowa Ruda, which also was the source of the sample used for curve 5, Figure 4. He made the very interesting suggestion that intermediate structural forms may exist between kaolinite and dickite, with interstratified groups of layers arranged as in these minerals, a suggestion very similar to the later ideas of Plançon and Tchoubar. The Jamaican samples have furnished an excellent series for testing this hypothesis, and (as we have seen) provide at least partial support. Keller *et al.* (1966) showed that the double endotherm of the Keokuk kaolinite, curve 6, changed towards the single peak and temperature range normally found for kaolinites when the Keokuk material was mechanically treated. They concluded that "perfection of crystallization is the predominant factor" determining the nature and temperature of the DTA endotherm.

An important respect in which the thermal behavior of dickites differs from that of kaolinites is the development of a reflection near 14A by some but not all dickites, and never by kaolinites (Hill, 1955; Roy and Brindley, 1956; Brindley, 1975). More recently, Brindley and Wan (1978) have shown that well-ordered dickites with sharp DTA endotherms give the best development of a 14A phase, and that less well-ordered dickites show little or no development of this phase.

From the combined X-ray and thermal data, the following conclusions are obtained. Structurally well-ordered dickites generally give sharp endotherms near 700°C and generate a 14A phase when dehydroxylation occurs. The dickite crystallization seems to be necessary because the exceptionally well-ordered Keokuk kaolinite does not give a 14A phase, although it gives a DTA endotherm resembling those of disordered dickites. Disordered dickites give mainly broad DTA endotherms and (from the previous study, Brindley and Wan, 1978) little or no development of the 14A phase. To this extent, a consistent picture of the structural and thermal properties of dickites emerges. However, there are exceptions to the general rule so that other variables, such as particle-size distribution, may be involved.

Other data pertaining to Jamaican dickites

Chemical and mineralogical data

The massive dickite samples, particularly from the northern vein at Job's Hill, show a variety of tints, mainly pink and pale green but also purple; less commonly they are white. Spectrographic analysis has yielded the data in Table 1 showing a much higher proportion of Ti in the pink material, and a higher proportion of Ca and Sr in the green material. These differences are consistent with their formation at different times by somewhat different solutions.

X-ray diffraction data show that crandallite, a hydrated calcium aluminum phosphate, is an impurity in all seven of the green samples and is practically absent from the four pink and purple samples. The higher Ca and Sr contents of the green material could arise from the crandallite impurity. We understand from Mr. G. H. Sabiston (private communication) that crandallite is a common impurity in Jamaican bauxites. We have no quantitative estimate of the proportion of crandallite in the green materials, but it is small. Anatase in the pink material is consistent with the relatively high Ti content.

Density data

A pycnometer determination of the density of the green material, using toluene as the liquid medium, gave the value 2.64, which is close to the density 2.60 calculated from the composition and unit-cell parameters of dickite. No particular significance is attached to the difference, 0.04, but the material is indeed dense, with no detectable porosity.

Conclusions

Dickites found at two localities near Job's Hill, Jamaica, exhibit various degrees of structural order-disorder and various DTA patterns. There is a tendency to pass from broad to sharp endotherms as structural order increases. Elsewhere it is shown that structurally ordered dickites tend to generate a 14A phase when dehydroxylation occurs. However, exceptions to these general observations do occur so that other factors besides structural order-disorder may influence the thermal behavior.

Table 1. Spectrographic analysis of pink- and green-tinted dickite samples from Job's Hill, Jamaica (Dr. N. H. Suhr, analyst)

	Pink	Green
Major elements	Al, Si	Al, Si
Ca	100 ppm	200 ppm
Sr	200	800
Ti	1000	10
Fe	50	30
Mg	10	10

Not detected in both samples: B, Be, Cr, Cu, Ag, Mn, Na, Y, Yb, Zr, Co, Cd, Zn, Sn, Bi, V, Pb, Ba.

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