

(The data are plotted against n , rather than t_n , in Fig. 1 as the shutter closed in between measurements and each measurement involved the same exposure of the crystal to the X-ray beam.) The initial calculation started with $S_0 = 2.24$, and after two cycles of refinement of S_n , along with the other 97 variables in the structure, the values of S_0 , a_1 , and a_2 had converged to $2.41(4)$, $-1.8(8) \cdot 10^{-4}$, and $-1.4(4) \cdot 10^{-7}$, with n , rather than t_n , the variable. Of more significance, the conventional R factor had converged to 13.3% for observations brought to a common scale through the use of the average change in the standards. An analysis of the weighted R factor as a function of $|F_0|$, $\lambda^{-1} \sin \theta$, and n indicated that there was a marked dependence on n , presumably because the use of the average change in the standards was an ineffective method of scaling the data. When the present method of scaling was used the calculation converged to an R value of 8.8%, although only two extra variables (a_1 and a_2) had been introduced. Moreover, the weighted R factor was no longer a function of n . Note that the plot of S_n/S_0 versus n derived by the present method differs markedly from the average curve for the four standards depicted in Fig. 1. This indicates that the average decomposition of these standards is not typical of the data set as a whole.

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Layer disorders in kaolinite during dehydration. By G. B. MITRA and S. BHATTACHERJEE, *Department of Physics, Indian Institute of Technology, Kharagpur, India.*

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An attempt has been made to explain the mechanism of the phase transformation of kaolinite to metakaolin by a quantitative study of the variation of g , the mean fractional change in the interlayer spacing in the $[00l]$ direction, and γ , the proportion of planes affected by such disorder, by application of the method developed by Mitra (*Z. Kristallogr.* (1963), 119, 165). γ and g values for samples heat treated at 500 and 600°C, for which only 002 reflexions were found to be tolerably sharp, were determined by a modified single-line technique. It is seen that the formation of metakaolin and the collapse of the kaolinite structure is preceded and accompanied by a disordering of the basal planes so that the interlayer spacing is variable.

Mitra (1958) and Brindley & Nakahira (1959) have observed that basal reflexions from kaolinite become asymmetrical as a result of dehydration at the temperature at which metakaolin starts to form. The asymmetry of the intensity profiles has been attributed by Mitra (1958) to irregularities in the interlayer distances of the basal planes. Mitra (1963) observed that this disorder exists even in ordinary kaolinite not subject to any heat treatment and has developed a method for quantitative determination of g , the mean fractional change in interlayer spacing, and of γ , the proportion of planes affected by such disorders. It is felt that the study of the variation of g and γ at different stages of the dehydration leading to the formation of metakaolin will ultimately be useful in understanding the mechanism of the phase transformation from kaolinite to metakaolin.

A sample of pure kaolinite from Georgia, U.S.A. was divided into several portions, each portion being separately heat treated in a muffle furnace for 10 hours at temperatures of 200, 300, 400, 500 and 600°C. Differential thermal analysis of the sample showed a faint endothermic peak at about 150°C and a more prominent endothermic peak at 562°C. X-ray diffraction line-profiles for various reflexions from the untreated as well as the dehydrated samples

Although in some of our data sets crystal decomposition has apparently affected the chosen standards equally though appreciably (Raymond, Corfield & Ibers, 1968), the present method would seem to be preferable to a correction derived from those standards since there is no assurance that other reflections are not affected at a different rate. Moreover, the present method would seem to be especially well suited for use with the procedure (Corfield, Doedens & Ibers, 1967) of collecting intensity data within a given angular range in 2θ effectively in random order.

The rapidity of convergence and the marked improvement in the agreement between $|F_0|$ and $|F_c|$ favor retention of the rather simple expression for S_n . Probably the introduction of additional terms in the power series would do little to improve the model further.

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were obtained with a Norelco X-ray diffractometer fitted with G.M. tube and decade counters. The technique of fixed count point to point recording of intensity was used and the accuracy achieved was about 1%. For the samples dehydrated at temperatures up to and including 400°C all basal reflexions present were broad and asymmetrical. The 005 and 006 reflexions were present only in the untreated samples. For these samples, the γ and g values for all 00/ reflexions were determined by the method described by Mitra (1963). For samples treated at 500 and 600°C only 001 and 002 reflexions were present; the 001 reflexions in both samples were very broad and diffuse while the 002 reflexions were less so. Hence the γ and g values in these samples were determined by adopting a single-line technique similar to that developed by Misra & Mitra (1967). For this purpose, the Fourier transform of the defect line-profile for the layer structure having a variable interlayer spacing in the $[00l]$ direction as derived, by Wilson (1962), has been calculated. The expression for the variance of the defect profile was then obtained from the formula developed by Wilson (1962). The resulting expression is given by

$$W_D = \frac{1}{4\pi^2} [2Sa - a^2], \quad (1)$$

where W_D is the variance of the disordered line profile, S the range over which the integration for the determination of W_D was made and $a = \gamma(1 - \cos 2\pi lg)$. The total variance (W) of the line profile, corrected for geometrical aberrations can now be written as

$$W = W_P + W_D,$$

where W_P is the variance resulting from the particle-size line profile. Proceeding as in Mitra (1964), we have for W , in 2θ units,

$$\frac{W \cos \theta}{\lambda S} = \frac{1}{\pi^2 P'} - \frac{a^2}{4\pi^2 d^2} - \frac{\lambda}{S \cos \theta}, \quad (2)$$

where $1/P' = 1/P + a/2d$; P is the particle size, d the interplanar distance; λ the wavelength of radiation used and θ the Bragg angle of reflexion. a was determined from the slope of the linear plot of $W \cos \theta / \lambda S$ versus $\lambda / S \cos \theta$. Knowing the values of a and Δ (the peak shift of the lines) γ and g values were calculated. The results are shown in

Table 1. Variation of g and γ with dehydration

Temperature (°C)	g	γ
30	0.148	0.063
200	0.171	0.065
300	0.170	0.080
400	0.173	0.077
500	0.210	0.033
600	0.218	0.021

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The crystal structure of ethylidene-*N,N*-diacetamide. BY B. KOLAKOWSKI *Institute of Physics, Polish Academy of Sciences, Warsaw, Poland*

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The crystal structure of ethylidene-*N,N*-diacetamide, $\text{CH}_3\text{CH}(\text{NHCOCH}_3)_2$, has been determined. The symmetry is monoclinic, space group $P2_1/c$. The molecules are linked by a double system of hydrogen bonds in layers parallel to (100) plane.

Ethylidene-*N,N*-diacetamide was prepared by the method described by Noyes & Forman (1933). Colorless crystals with melting point 180°C were grown by slowly evaporating a concentrated methyl alcohol solution. The crystal belongs to the monoclinic system with the space group $P2_1/c$. The unit cell has dimensions $a = 8.52$; $b = 9.54$; $c = 9.57$ Å; $\beta = 92^\circ 47'$ determined from Weissenberg and oscillation photographs taken about a , b and c axes with nickel-filtered $\text{Cu } K\alpha$ radiation. There are four molecules per unit cell, all in general positions. Intensities were measured by visual estimation from the equi-inclination Weissenberg photographs with series of calibrated spots. The number of independent reflexions was 674, of which 585 were in the measurable range. Lorentz-polarization corrections were made. Absorption corrections were considered to be unnecessary because of the small crystal used. The preliminary structure has been deduced from a geometrical consideration and Patterson synthesis. A three-dimensional least-squares refinement of the 30 positional parameters and 10 individual isotropic temperature parameters of carbon, nitrogen and oxygen atoms was carried out. After several cycles of refinement the value of R decreased to 0.14. Table 1 lists the final fractional coordinates and the thermal param-

eters for all the atoms of C, N and O. The projection of the structure along [010], and bond lengths and bond angles, are shown in Figs. 1 and 2 respectively. The average standard deviations in these are 0.015 Å in the bond length and about 1° in the bond angle.

It is evident from Table 1 that with increasing dehydration at 200°C the mean fractional change in the interlayer distances of the basal planes increases considerably (from 0.148 to 0.171) while the proportion of planes involved remains almost unaffected (0.063 to 0.065). At 300°C more planes are affected - γ changing from 0.065 to 0.080; however, g remains constant. At 400°C , both γ and g are constant, but at 500°C there is a large increase in g while γ has considerably decreased. This shows that collapse of the kaolinite structure has started - the larger proportion of the already affected planes having been converted to meta-kaolin. Only a small proportion of the planes in the kaolinite structure remain but with greater disorder. This process is continued at 600°C . Thus the formation of meta-kaolin and collapse of the kaolinite structure is preceded by and accompanied by disorder of the basal planes so that the interlayer spacing is variable.

Details of this work will soon be published elsewhere.

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Table 1. Final fractional coordinates and thermal parameters

	x/a	y/b	z/c	B
C(1)	0.244	0.520	0.015	2.93
C(2)	0.307	0.388	0.091	2.58
C(3)	0.088	0.222	0.080	2.18
C(4)	0.036	0.139	0.155	3.61
C(3')	0.522	0.348	0.262	3.17
C(4')	0.639	0.405	0.374	3.76
N(5)	0.179	0.309	0.154	2.03
N(5')	0.420	0.432	0.208	2.41
O(6)	0.107	0.199	0.051	4.17
O(6')	0.535	0.219	0.223	4.62

The molecules in the crystal lattice are linked together by two series of hydrogen bonds. One series of length 2.96 Å runs approximately parallel to the b axis and the other of length 2.88 Å runs approximately parallel to the c axis.