# *An X-ray method for studying orientation of micaceous minerals in shales, clays, and similar materials.*

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## 1. *Introduction.*

MICACEOUS minerals, such as the micas, the chlorites, and most clay minerals, usually develop as thin lamellae parallel to the crystallographic basal plane (001). In sedimentary deposits these lamellae tend to lie parallel to the bedding plane and in slates parallel to the cleavage. Their degree of orientation is likely to vary with the conditions of formation of the material, and in the case of slates Bates (1947) has shown that it is closely related to their fissility. In relatively coarse-grained materials the degree of orientation may be studied directly by suitable microscopic techniques, but with fine-grained materials X-ray methods must be used, and even with coarser-grained materials the use of X-ray methods may have advantages, especially if a scanning technique, such as that described by Thewlis and Pollock (1950), is employed.

The orientation of micaceous minerals is most easily investigated by considering the intensities of X-rays reflected from their basal (001) planes in different directions with respect to the bedding or cleavage plane of the material. With X-ray powder cameras of conventional type in which the reflected X-rays are recorded through an aperture on a strip of film, a series of photographs for different angular settings of the material will be required. If, however, a complete Debye-Scherrer diagram is recorded, then a single photograph taken with one angular setting of the material suffices to give the required information.

Custers (1948) has analysed in detail this method for studying orientation with particular reference to rolled metal sheets. The practical problems are somewhat different with micaceous materials, since the fragility of many specimens makes the use of small blocks of material almost essential and these can be studied only by surface reflection methods. With thin sheets both surface reflection and transmission arrangements may be employed. With metals more penetrating radiations of shorter wave-length can be usefully used than are practicable

with micaceous minerals, since their low-order basal reflections are mainly used. MacEwan and Norrish (1951) have also considered the effect of orientation on the intensities of X-ray diffractions from clays, but their analysis, which has so far been published only in abstract, appears to be more general than is essential for the study of orientation alone. The present paper is restricted to the study of orientation in blocks of micaceous materials by a surface reflection method. The analysis differs somewhat from that given by Custers, but the same final expressions are obtained.

## 2. *General description of the method.*

If OP (fig. 1) represents the normal to a lamella surface or basal plane and ON the normal to the plane of cleavage or bedding, then the orientations of the lamellae can be expressed by the distribution function, $p$, of the normals OP with respect to ON. If the angle $\mathrm{P\hat{O}N}$ is denoted by $\xi$, then $p$ will usually be a function only of $\xi$; but if necessary a second angular variable $\chi$ can be introduced, giving the variation of $p$ around the normal ON. $p(\xi)$ can be defined as the number of normals, OP, per unit solid in angle in direction $\xi$, and $p(\xi, \chi)$ the number in direction $(\xi, \chi)$.

In fig. 1, the incident X-ray beam lies along the Y-axis of the rectangular co-ordinate system OX, OY, OZ. The specimen block is placed with its natural cleavage face containing the Z-axis and with surface normal ON lying in the plane XOY and making an angle $\alpha$ with OX. The photographic plate, SYT, lies parallel to XOZ. The arc SRT represents a portion of a Debye-Scherrer ring corresponding to a Bragg angle $\theta$, so that $\mathrm{R\hat{O}Y} = 2\theta$. Then OP, the normal to the reflection plane, bisects $\mathrm{R\hat{O}Y'}$. As R sweeps over the arc SRT, OP generates a conical surface with a semi-vertical angle $(90-\theta)$.

Consider now a sphere of unit radius surrounding O with contours of equal $p(\xi)$ or $p(\xi,\chi)$ on its surface. If $p$ is a function of $\xi$ only, as we may generally assume (see, however, § 7), the contours will be circles concentric with ON. The X-ray intensity recorded at a point R, with co-ordinates $(r, \psi)$ on the photographic plate, will depend (inter alia) on the value of $p$ at the point P. As R moves round the Debye-Scherrer ring, P traces on the sphere a circle lying parallel to the plane XOZ, the path of which in relation to the $p$-contours determines what information concerning $p$ is obtainable from a single Debye-Scherrer ring. The most complete information is obtained when P passes through N, and this requires that $\alpha = \theta$. The powder block is then set symmetrically with respect to the incident and reflected X-rays in the horizontal plane.

If we consider now a small range of directions $\Delta\theta$ for the incident radiation so that the Debye-Scherrer ring has a finite radial width, and consider also a small circumferential element $r\Delta\psi$, then the corresponding area in which P lies on the unit sphere will be $\Delta\theta\Delta\xi$ where $\Delta\xi$ is the movement of P corresponding to $\Delta\psi$. The number of planes reflecting

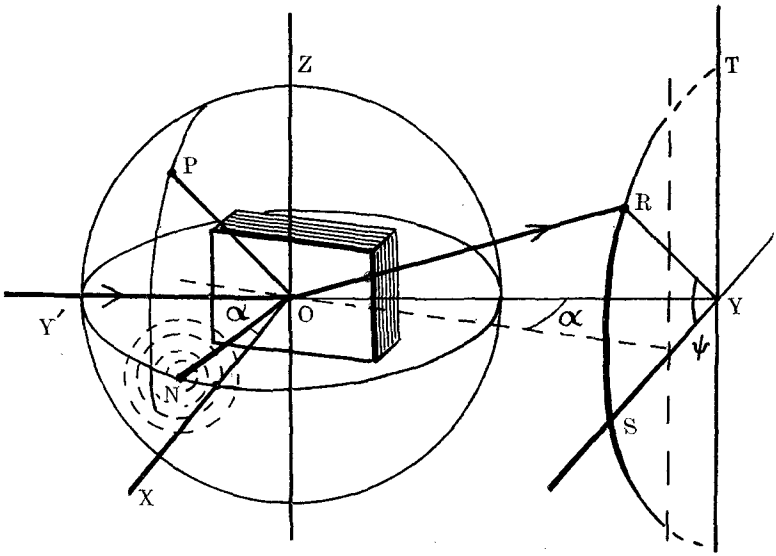


FIG. 1. To illustrate X-ray reflection from a flat block of fine-grained material. Y'O = incident beam; OR = typical reflected beam; OP = normal to reflecting plane; SRT = Debye-Scherrer arc; ON = normal to block surface; $N\hat{O}P = \xi$. Dashed lines surrounding N represent contours of $p(\xi)$.

into the small radial segment of the Debye-Scherrer ring will therefore be $p(\xi)\Delta\theta\Delta\xi$ and the reflected intensity will be given by

$$I(\psi)\Delta\psi \propto p(\xi)\Delta\theta\Delta\xi A(\alpha, \theta, \psi) \qquad (1)$$

where $A(\alpha, \theta, \psi)$ is the absorption factor appropriate to the conditions specified by $(\alpha, \theta, \psi)$. To obtain $p(\xi)$ from measurements of $I(\psi)$, two calculations are required, viz. $A(\alpha, \theta, \psi)$ and the relation of $\Delta\xi$ to $\Delta\psi$.

### 3. *The absorption factor*, $A(\alpha, \theta, \psi)$.

X-ray absorption in flat powder blocks has been analysed and discussed by Brentano (1927, 1935) and by Brindley and Spiers (1934), and although they were then concerned primarily with rays such as OS

in the equatorial plane (fig. 1), their analysis is equally applicable to the present problem and the same expression is obtained for A, namely

$$A = \frac{1}{\mu} \frac{\sin \beta}{(\sin \alpha + \sin \beta)}$$

where $\alpha$ and $\beta$ are the glancing angles of incidence and of emergence of the X-rays with respect to the surface of the block and $\mu$ is the linear absorption coefficient. The integration involved in obtaining A presupposes that the block is sufficiently thick to absorb fully the incident radiation.

The angle $\beta$ is the complement of $N\hat{O}R$ in fig. 1 and is calculated as follows:

The direction-cosines of ON and OR are respectively,

ON, $\cos \alpha, -\sin \alpha, 0$;

OR, $\sin 2\theta \cos \psi, \cos 2\theta, \sin 2\theta \sin \psi$.

Then $\cos N\hat{O}R = \sin \beta = \sin 2\theta \cos \psi \cos \alpha - \cos 2\theta \sin \alpha$

Whence
$$A = \frac{1}{\mu} \left( \frac{\sin 2\theta \cos \psi \cos \alpha - \cos 2\theta \sin \alpha}{\sin 2\theta \cos \psi \cos \alpha - \cos 2\theta \sin \alpha + \sin \alpha} \right) \tag{2}$$

and for the symmetrical setting of the block, when $\alpha = \theta$,

$$A = \frac{1}{2\mu} \left( \frac{1 - 2\cos^2\theta (1 - \cos \psi)}{1 - \cos^2\theta (1 - \cos \psi)} \right) \tag{2a}$$

These results agree with formulae given by Custers (1948b).

### 4. *The relation of $\xi$ to $\psi$.*

The direction cosines of OP are obtained by using the fact that OP bisects $R\hat{O}Y'$, and are $\cos \theta \cos \psi$, $-\sin \theta$, $\cos \theta \sin \psi$. This result, combined with that for ON, gives

$$\cos P\hat{O}N = \cos \xi = \cos \theta \cos \psi \cos \alpha + \sin \theta \sin \alpha \tag{3}$$

and for the symmetrical setting,

$$\cos \xi = \cos \psi \cos^2\theta + \sin^2\theta \tag{3a}$$

or, for small values of $\theta$,

$$\xi \doteqdot \psi \text{ and } \Delta\xi \doteqdot \Delta\psi \tag{3b}$$

Actually $\theta$ need not be very small in order that this approximation shall be a good one, as the data in table I show. For $\psi$ in the range 0°–60° (the following section shows that this is the full angular range available) $\xi$ does not differ appreciably from $\psi$ provided $\theta$ is less than about 20°; for micaceous minerals, $\theta$ is likely to be about 10° or less, and the condition is easily satisfied.

TABLE I. Values of $\xi$ for a range of values of $\psi$ and $\theta$.

| $\theta$ \ $\psi$ | 0. | 10. | 20. | 30. | 40. | 50. | 60°. |
|---|---|---|---|---|---|---|---|
| 10° | 0 | 9·7 | 19·7 | 29·5 | 39·4 | 49·2 | 59·0 |
| 20 | 0 | 9·2 | 18·7 | 28·1 | 37·5 | 46·8 | 56·0 |
| 30 | 0 | 8·5 | 17·3 | 25·9 | 34·5 | 42·9 | 51·3 |

### 5. *The maximum angular range of investigation of $\xi$ and $\psi$.*

The angular range of $\psi$, and therefore of $\xi$, which can be studied is limited by the shadow cast by the specimen block on the photographic plate. It is seen that

$$\cos \psi_{\max} = \tan \alpha / \tan 2\theta$$

and for the symmetrical setting,

$$\cos \psi_{\max} = \tan \theta / \tan 2\theta = 1/2$$

or

$$\psi_{\max} \doteqdot 60°.$$

The range of orientations which can be studied usefully by this method is somewhat less than 60° because as $\psi \to \psi_{\max.}$ so $A \to 0$. The useful range of investigation is therefore for values of $\xi$ up to about 50°. Alternatively, by combining equations (2a) and (3a), we find

$$A = (2 - \sec \xi)/2\mu$$

so that $A$ becomes zero when $\xi = 60°$.

### 6. *The distribution of $I(\psi)$ in a Debye-Scherrer ring and the determination of $p(\xi)$.*

The required result is obtained by combining equations (1), (2a), and (3b) and omitting constant factors:

$$p(\xi) \propto I(\psi) \left( \frac{1 - \cos^2\theta\,(1 - \cos \psi)}{1 - 2\cos^2\theta\,(1 - \cos \psi)} \right)$$

If $p(0)$ and $I(0)$ are values corresponding to $\xi = 0$ and $\psi = 0$, then

$$\frac{p(\xi)}{p(0)} = \frac{I(\psi)}{I(0)} \left( \frac{1 - \cos^2\theta\,(1 - \cos \psi)}{1 - 2\cos^2\theta\,(1 - \cos \psi)} \right) \tag{4}$$

By careful photometry of the Debye-Scherrer arc, $I(\psi)$ can be obtained and hence, by means of equation (4), the variation of $p(\xi)$ with $\psi$, i.e. with $\xi$.

### 7. *The case when p is a function of $\xi$ and $\chi$.*

It can readily be determined whether $p$ is a function solely of $\xi$ or of two variables $\xi$ and $\chi$ by taking further X-ray photographs after turning the specimen block through an angle $\chi$ about the surface normal, *ON*. In this way the variation of $p(\xi, \chi)$ with respect to $p(0)$ can be determined within the limitation that $\xi$ cannot exceed about 60°.

### 8. *Some representative X-ray orientation diagrams.*

The present paper is concerned primarily with the method and it is not intended to discuss detailed photometric analyses of X-ray photographs. Fig. 2 shows six representative photographs of clays and shales taken with Cu-$K\alpha$ radiation and with the specimen block set at $\alpha = 12\frac{1}{2}°$ in order to satisfy reasonably well the condition $\alpha \quad \theta$ for both the (002) basal reflection from kaolinite and the adjacent $(10\bar{1}1)$ reflection from quartz, these reflections being indicated in the photographs. Since there is no evidence for any preferential crystallographic orientation of the quartz particles, one obtains directly a qualitative picture of the degree of orientation of the kaolinite particles by comparing visually the intensity variations along these two arcs.

After each X-ray exposure the specimen block was removed and the direct beam recorded; this is seen in the lower left-hand corner of each diagram. Each photograph is one half of the original and corresponds to values of $\psi$ above the axis $SY$ in fig. 1. The direction $\psi = 0$ lies along the lower edge of each diagram. The shadow cast by the specimen block is clearly seen and the maximum value of $\psi$ is about 60°.

The photographs in fig. 2 are as follows:

(*a*) A Coal Measure shale, taken from 8 feet above the Haigh Moor Coal, from a borehole core at Wentbridge, Yorks. This shale splits moderately easily. X-rays were reflected from the bedding-plane surface. The majority of the kaolinite particles are seen to be within about $\pm 20°$ of this surface, as shown by the radial line in fig. 2*a* which corresponds to $\psi = 20°$.

(*b*) The same Coal Measure shale. X-rays were reflected from a ground surface at right angles to the bedding. The (002) kaolinite arc is now practically invisible.

(*c*) A well-laminated Coal Measure shale taken from 25 feet below a thin coal band, Robin Hood quarry, near Wakefield, Yorks. This shale

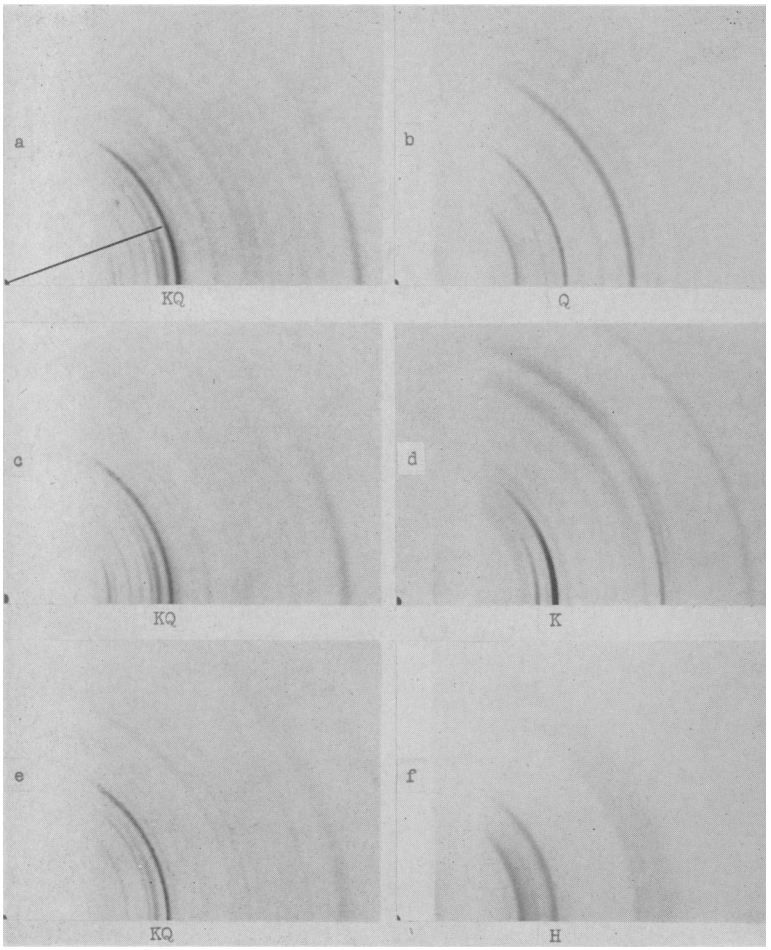


FIG. 2. X-ray orientation diagrams of shales and clays. (*a*), (*b*), and (*c*), Coal Measure shales. (*d*), Cornish china-clay. (*e*), Coal Measure 'clunch'. (*f*), Utah halloysite, dehydrated at 300° C.

Q = Quartz ($10\bar{1}0$), K = Kaolinite (002), H = Halloysite (002).

splits very readily into flakes with smooth, flat surfaces. The orientation is somewhat better than in (*a*).

(*d*) A pressed block of Cornish china-clay containing no quartz. The

surface had a polished appearance. The orientation is less perfect than in (*a*) and (*c*), which is contrary to what was anticipated.

(*e*) A Coal Measure 'clunch', i.e. a shaly material, not well bedded, taken from a borehole core at Wentbridge, Yorks., 40 feet above the Flockton Thick Coal. The orientation is clearly much less perfect than in (*a*) and (*c*).

(*f*) A compressed block of Utah halloysite, dehydrated at 300° C. This shows no orientation as would be expected from the known morphology of this mineral, which consists of rod- and tube-like particles rather than flakes.

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