(iv) Diffraction from specimens in controlled environments (*e.g.* pressure cells) with restricted apertures in which the time taken to collect data conventionally may be excessive. The forward-scattered beams in the Laue method tend to be of very short wavelengths (on the SRS wiggler beam the short wavelength cut-off will be 0·2 Å) and hence are only weakly absorbed by the sample container.

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Chlorite Polytypism.
I. OD Interpretation and Polytype Symbolism of Chlorite Structures

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Abstract

In order to establish identification criteria for chlorites, all their polytypes with maximum degree of order (MDO polytypes, identical with 'standard', 'regular' or 'simple' polytypes) have been derived. There are 164 di-dioctahedral, 44 tri-dioctahedral and 20 tri-trioctahedral MDO polytypes. The geometrical analysis leading to their derivation was based on the idealized model of the chlorite structures which can be interpreted as an order–disorder (OD) structure. This made it possible to apply consistently the apparatus of the OD theory. Individual polytypes are described by fully descriptive symbols – a comparison with the symbols of other authors is also given.

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1. Introduction

This paper is a continuation of a series dealing with polytypism of sheet silicates from the point of view of the theory of order–disorder (OD) structures (Dornberger-Schiff, 1964, 1966, 1979; Dornberger-Schiff & Fichtner, 1971). Practical applications of the results to these substances appeared, for example, in the papers of Mikloš (1975), Weiss & Mikloš (1979), Weiss & Ďurovič (1980). The important definitions and concepts of the OD theory (as applied to sheet silicates) were given in some detail by Dornberger-Schiff & Ďurovič (1975*a*), to which the reader who is not familiar with these ideas is particularly referred. A unified system of fully descriptive polytype symbols for the main types of sheet silicates has also been worked out (Ďurovič & Dornberger-Schiff, 1979).

The structures of chlorites and vermiculites have very much in common (Weiss & Ďurovič, 1980). The reader is thus asked to consult the paper on vermiculites (WĎ in the following) for basic terms and definitions.*

2. OD interpretation of the idealized model of chlorite structures

Any of the two octahedral sheets in chlorite structures may possess one of the following three symmetries: $H(\bar{3})12/m$, $P(\bar{3})12/m$, $P(3)12$, depending on whether the three octahedral positions per unit mesh are occupied by three equal, two equal and one different, or three different crystallochemical entities (atoms or voids), respectively. The third case has not been encountered in chlorites as yet and we shall exclude it from our further considerations. The first and the second case correspond as a rule to trioctahedral and dioctahedral species, respectively, and we shall refer to them as such.* Keeping this in mind, we arrive at the following choice of OD layers for which any chlorite structure can be qualified as an OD structure (six OD layers per repeat unit, Fig. 1).

L_{6n} OD layer (denoted Ic in the following) containing a plane of octahedrally coordinated interlayer cations and halves of the two adjacent planes of interlayer OH⁻ ions; layer symmetry $H(\bar{3})12/m$ (trioctahedral), $P(\bar{3})12/m$ (dioctahedral); origin at any octahedral position (tri-), or at a void (di-).

$L_{6n\pm1}$ OD layer (denoted OH) containing the planes of the remaining halves of interlayer OH⁻ ions; symmetry $H(6)mm$, origin at hexad.

$L_{6n\pm2}$ OD layer (denoted Tc) containing basal O atoms, tetrahedral cations, and halves of the apical O atoms and of the OH⁻ ions at their level − all belonging to the tetrahedral sheet of the 2:1 layer; symmetry $P(6)mm$, origin at hexad.

$L_{6n\pm3}$ OD layer (denoted Oc) containing a plane of octahedrally coordinated cations and halves of the adjacent planes of apical O atoms and of OH⁻ ions at their level; symmetry and origin as for L_{6n}.

For the same reason as for vermiculites it follows that any chlorite structure consists of four kinds of OD layers (provided that Ic and Oc keep their character throughout the structure), belongs to the category IV and has the following OD groupoid family symbol (Grell & Dornberger-Schiff, 1982):

Symm Ic　　$H(6)mm$　　　　$P(6)mm$　　Symm Oc

$[\tfrac{1}{3},0]$　　　$[-\tfrac{1}{3},-\tfrac{1}{3}]$　　　$[\tfrac{1}{3},0]$

where Symm Ic and Symm Oc stand for the layer group of Ic and Oc, respectively. Accordingly, it is usual (e.g. Drits & Karavan, 1969; Zvyagin, Vrublevskaya, Zhoukhlistov, Sidorenko, Soboleva & Fedotov, 1979) to distinguish di-dioctahedral (dioctahedral for short), di-trioctahedral, tri-dioctahedral and tri-trioctahedral (trioctahedral for short) chlorite families. The first prefix refers to the 2:1 layer, the second to the interlayer hydroxide sheet.

From the symmetry of the individual OD layers and one of their possible relative positions to the preceding layer as indicated in the symbol of the OD groupoid family, the number of positions of an OD layer relative to its predecessor, leading to geometrically equivalent layer pairs [the so-called NFZ relations, see WĎ, equation (2)], can be calculated.*

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* See deposition footnote.

PACKETS	BL's	STRUCTURAL SCHEME	OD LAYERS		POLARITY OF OD LAYERS
	B_5		L_{10}		
P_2	B_4		L_9		
			L_8		
	B_3		L_7		
			L_6		
P_1			L_5		
	B_2		L_4	Tc	polar
			L_3	Oc	nonpolar
P_0	B_1		L_2	Tc	polar
			L_1	OH	polar
	B_0		L_0	Ic	nonpolar
			L_{-1}	OH	polar
P_{-1}	B_{-1}		L_{-2}		
			L_{-3}		

Fig. 1. Schematic representation of a chlorite structure showing OD layers, their labelling, OD packets, building layers (BL's), and the polarity of the OD layers.

Evidently, Mg-vermiculite can be treated as if it belonged to the tri-dioctahedral family, because the symmetries of Ic and Oc, respectively, are the same, no matter whether two of the three octahedral positions of Ic are occupied and the third is empty, or *vice versa*. As in vermiculites, only the nine displacement vectors $\mathbf{v}_{pq}(q - p \geq 1)$ shown in Fig. 2, from L_p to L_q, may occur in chlorite structures.

OD packets

OD packets (Ďurovič, 1974) constitute the basis for the derivation of MDO polytypes; any packet consists of one half of the 2:1 layer (2:1 half) and of the adjacent half of the interlayer hydroxide sheet (interlayer half). To derive all kinds of OD packets for any of the four chlorite families, we have to consider the individual symmetry of either of these two halves and their relative positions which are in keeping with the possible relative positions of OD layers. For the dioctahedral family any 2:1 half has the symmetry Cm and it can appear in six different orientations labelled 0 to 5 depending on $\mathbf{v}_{6n-3,6n-2}$ or $\mathbf{v}_{6n+2,6n+3}$. We choose its origin at the hexad of $L_{6n\pm2}$. Any interlayer half has the symmetry $P(3)1m$ and it can appear only in two orientations e and u depending on the orientation of L_{6n}. The displacement vector between these two halves can be any of the nine displacement vectors shown in Fig. 2 with the proviso (*cf.* WĎ, p. 636) that a displacement vector with uneven (even) character is compatible only with the e (u) orientation of the interlayer half, whereas a displacement vector $\langle*\rangle$, $\langle+\rangle$ or $\langle-\rangle$ is compatible with either orientation. Accordingly, for a fixed orientation of the 2:1 half, we obtain $3 + 3 + 2 \times 3 = 12$ non-congruent (8 non-equivalent) kinds of di-dioctahedral packets. For tri-dioctahedral packets (as in vermiculites) 8 non-congruent (6 non-equivalent) kinds of packets result, since their 2:1 half has higher $[P(3)1m]$ symmetry.
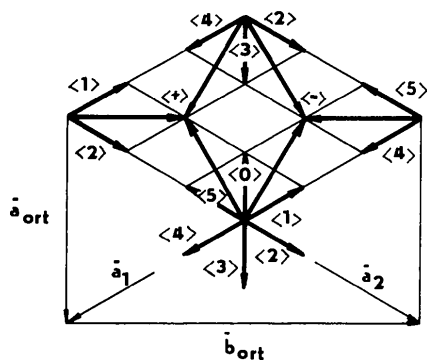
The interlayer half in di-tri- and tri-trioctahedral families has the symmetry $H(3)1m$; its unit mesh is triply primitive compared with the P mesh of the 2:1 half. Its three translationally equivalent origins can thus be reached from the origin of the 2:1 half *simultaneously* by three displacement vectors, and it is easy to see that only the following three combinations can occur: $\langle0\rangle \wedge \langle2\rangle \wedge \langle4\rangle$, $\langle1\rangle \wedge \langle3\rangle \wedge \langle5\rangle$ or $\langle*\rangle \wedge \langle+\rangle \wedge \langle-\rangle$ which will be designated e, u or x, respectively, in the following. Displacements e (u) are compatible only with the u (e) orientation of the interlayer half; displacements x are compatible with both orientations. Thus for the tri-trioctahedral family, four kinds of OD packets with symmetry $P(3)1m$ result and the same number results also for the di-trioctahedral family (symmetry Cm). The results of the above considerations are summarized in Table 1.

We may note that in all packets in which one of the displacements $\langle*\rangle$, $\langle+\rangle$, $\langle-\rangle$ (or $\langle x\rangle$) occurs, the interlayer cations coincide in the normal projection onto the $X_1 X_2$ plane, with the tetrahedral cations of the adjacent 2:1 half.

OD packets are by definition polar and any chlorite structure consists of even-numbered and odd-numbered packets (Fig. 1) with a regular alternation of their sense of polarity in the stacking direction. Two adjacent packets correspond to one *unit structure* defined according to the AIPEA recommendation (Bailey, 1980). This should be kept in mind when comparing the results of this paper with those of Drits & Karavan (1969) or of Zvyagin *et al.* (1979), where six types of 'packets' are mentioned. In fact, the Russian term 'пакет' means unit structure and it has been wrongly translated into English as 'packet'. On the other hand, it is usual in the literature to speak of chlorites containing n unit structures per identity period as n-layer polytypes and we shall also use this designation in the following.

3. Polytype symbols and MDO polytypes

The stacking sequence in any chlorite polytype can be described by fully descriptive symbols which – for families with a dioctahedral interlayer – have the same appearance as those for Mg-vermiculite (*cf.* WĎ, p. 636):

$$T_0 \quad T_1 \ . \ T_2 \quad T_3 \quad T_4 \ . \ T_5 \quad ...,$$
$$\quad\ v_{01} \quad v_{12} \quad v_{23} \quad v_{34} \quad v_{45}$$

where the characters refer to *building layers* (BL's): $B_{3n\pm1}$ for the two halves of the 2:1 layer, B_{3n} for the entire interlayer (Fig. 1). The symbols for the families with a trioctahedral interlayer necessitate an additional displacement character $v_{3n-1,3n+1}$ placed in the third line below T_{3n} since the unit mesh of B_{3n} is triply primitive, the displacement characters $v_{3n-1,3n}$ and $v_{3n,3n+1}$ can

Fig. 2. Displacement vectors $\langle v\rangle$, their relation to the basic vectors $\mathbf{a}_1$, $\mathbf{a}_2$ and their conventional characters: $\langle0\rangle = (-\frac{1}{3},-\frac{1}{3})$; $\langle1\rangle = (-\frac{1}{3},0)$; $\langle2\rangle = (0,\frac{1}{3})$; $\langle3\rangle = (\frac{1}{3},\frac{1}{3})$; $\langle4\rangle = (\frac{1}{3},0)$; $\langle5\rangle = (0,-\frac{1}{3})$; $\langle*\rangle = (0,0)$; $\langle+\rangle = (-\frac{1}{3},\frac{1}{3})$; $\langle-\rangle = (-\frac{1}{3},\frac{1}{3})$. The orthogonal basic vectors are also shown.

Table 1. *OD packets of four families of chlorite polytypes, their symmetry and the relations of isomorphy and homomorphy between them*

Any packet is described by the orientational characters of its two constituting parts separated in the lower line by a displacement character. Only odd-numbered images of symbols of packets in their standard orientation are listed.

Di-trioctahedral		Di-dioctahedral		Tri-dioctahedral		Tri-trioctahedral	
Packet symbol	Layer group	Packet symbol	Layer group	Packet symbol	Layer group	Packet symbol	Layer group
		.0 u / *	$C1m(1)$	.e u / *	$P(3)1m$		
.0 u / x	$C1m(1)$	.0 u / + ⎫ .0 u / − ⎬	$P1$	.e u / + ⎫ .e u / − ⎬	$P(3)$	.e u / x	$P(3)1m$
		.0 e / *	$C1m(1)$	.e e / *	$P(3)1m$		
.0 e / x	$C1m(1)$	.0 e / + ⎫ .0 e / − ⎬	$P1$	.e e / + ⎫ .e e / − ⎬	$P(3)$	.e e / x	$P(3)1m$
		.0 u / 0	$C1m(1)$				
.0 u / e	$C1m(1)$	.0 u / 2 ⎫ .0 u / 4 ⎬	$P1$	.e u / 0	$C1m(1)$	.e u / e	$P(3)1m$
		.0 e / 3	$C1m(1)$				
.0 e / u	$C1m(1)$	.0 e / 1 ⎫ .0 e / 5 ⎬	$P1$	.e e / 3	$C1m(1)$	.e e / u	$P(3)1m$
4 non-congruent		12 non-congruent 8 non-equivalent		8 non-congruent 6 non-equivalent		4-non-congruent	

only be *e*, *u*, or *x*, so that the sum of the corresponding displacement vectors is ambiguous. It has thus to be indicated in the symbol which concrete path from B_{3n-1} to B_{3n+1} is realized. The symbol then acquires the form

$$T_0 \quad T_1 \ . \ T_2 \quad T_3 \quad T_4 \ . \ T_5 \quad T_6$$
$$v_{01} \quad v_{12} \quad v_{23} \quad v_{34} \quad v_{45} \quad v_{56} \quad \dots$$
$$v_{\bar{1}1} \qquad\qquad v_{24} \qquad\qquad v_{57}$$

The smallest repeating part of the periodic sequence of characters, placed between vertical bars (|...|), indicates a periodic polytype.

The characters in the above symbols are not mutually independent. The *limiting parity conditions* which hold for their compatibility are summarized in Table 2.

The symbols constructed in this way* made it possible to include chlorites and vermiculites in one unified notation system with other sheet silicates (Dornberger-Schiff & Ďurovič, 1975*b*; Ďurovič & Dornberger-Schiff, 1976, 1978) and comply with the requirements for fully descriptive polytype symbolisms (Bailey *et al.*, 1977). They enable the user to recognize relevant crystallochemical and geometrical features of any chlorite polytype: the family to which it belongs,

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* See deposition footnote.

Table 2. *Parity conditions limiting the compatibility of the characters in chlorite polytype symbols*

	Dioctahedral						Trioctahedral	
$T_{3n-2}\cdot T_{3n-1}$ compatible only with	0·0	0·4	0·2	1·1	1·5	1·3		
	2·4	2·2	2·0	3·5	3·3	3·1	*e.e*	*u.u*
	4·2	4·0	4·4	5·3	5·1	5·5	(redundant)	
↓								
$v_{3n-2,3n-1}$	3	5	1	4	0	2		
			(redundant)				1,3,5	0,2,4
T_{3n} compatible only with	*e*		*u*				*e*	*u*
↓								
$v_{3n-1,3n}$ $v_{3n,3n+1}$	{ 1,3,5 *,+,−		0,2,4 *,+,− }				*u,x*	*e,x*

Moreover for a trioctahedral interlayer

T_{3n} $v_{3n-1,3n} v_{3n,3n+1}$ compatible only with	$\dfrac{e}{u}\ \dfrac{u}{u}$ or $\dfrac{u}{e}\ \dfrac{e}{e}$	$\dfrac{e}{x\ x}$ or $\dfrac{u}{x\ x}$	$\dfrac{u}{e\ e}$ or $\dfrac{u}{e\ e}$
↓	$\dfrac{u}{e\ x}$ or $\dfrac{u}{x\ e}$		$\dfrac{e}{u\ x}$ or $\dfrac{e}{x\ u}$
$v_{3n-1,3n+1}$	0,2,4	*,+,−	1,3,5

the periodicity, the structure of the 2:1 layer (centrosymmetric or noncentrosymmetric), the position and orientation of the interlayers, facilitate its pictorial representation, determination of its symmetry (Ďurovič

& Dornberger-Schiff, 1979) and calculation of its Fourier transform. We may show four typical examples:

(1) di-dioctahedral $\begin{vmatrix} u & 0.0 & u & 0.0 \\ + & 3 & - & - & 3 & + \end{vmatrix}$ two-

layer (four-packet) polytype;

(2) di-trioctahedral $\begin{vmatrix} e & 2.4 & e & 4.2 \\ x & 3 & x & x & 3 & x \\ * & & & * & \end{vmatrix}$ two-layer

(four-packet) polytype;

(3) tri-dioctahedral $\begin{vmatrix} e & e.e & e & e.e & e & e.e \\ 5 & 3 & 1 & 1 & 5 & 3 & 3 & 1 & 5 \end{vmatrix}$

three-layer (six-packet) polytype;

(4) tri-trioctahedral $\begin{vmatrix} u & e.e \\ e & 3 & e \\ 3 & & \end{vmatrix}$ one-layer (two-

packet) polytype.

The polytype (1) contains centrosymmetric and (2) noncentrosymmetric 2:1 layers. Interlayer cations project onto the tetrahedral cations of the adjacent 2:1 layers in (1) and (2); this is not the case in (3) and (4). Octahedral sheets in the interlayers and in the 2:1 layers are parallel in (2) and (3) (the same parity of the corresponding orientational characters) and antiparallel in (1) and (4).

A generalization of the symbolism for families with hetero-octahedral sheets, similar to those for micas (Dornberger-Schiff, Backhaus & Ďurovič, 1982), is also possible.

Relation to other symbolisms

So far, two fully descriptive symbolisms for chlorites have been used in the literature. The first system was proposed initially for one-layer trioctahedral polytypes by Brown & Bailey (1962) and later generalized for all chlorite polytypes by Lister & Bailey (1967). The second system was elaborated by Zvyagin (1967, 1974) and Zvyagin et al. (1979). A comparison of these two systems with our system as well as a conversion table between all three systems together with an example have been deposited.*

MDO polytypes

There is an infinite number of periodic as well as aperiodic polytypes within any family. In order to select periodic polytypes including those which are most likely to occur in natural or synthetic samples, the concept of MDO polytypes has been used

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* See deposition footnote.

(Dornberger-Schiff & Grell-Niemann, 1961; Dornberger-Schiff, 1982; Dornberger-Schiff & Grell, 1982). The MDO criteria applied to chlorite structures single out periodic polytypes which consist of equivalent OD packets, contain two regularly alternating kinds of pairs of adjacent OD packets, and in which all triples of consecutive OD packets are equivalent. This is in accordance with the general philosophy of the building of crystal structures: a configuration which is more stable than other configurations will occur alone (if this is possible at all) and not intermixed with others.

The MDO polytypes for the dioctahedral family have been derived by using the same procedure as for Mg-vermiculite (WĎ, p. 637); trioctahedral MDO polytypes have been obtained from these by forming homomorphous series and selecting their trioctahedral representatives. The MDO polytypes for the tri-dioctahedral family are formally identical with those for Mg-vermiculite. The di-trioctahedral family has not been handled as yet – this problem will be dealt with in due course.

All in all there are 164 non-equivalent (256 non-congruent) di-dioctahedral, 44 (60) tri-dioctahedral and 20 (24) tri-trioctahedral MDO polytypes.*

Concluding remark

The majority of chlorites found so far are trioctahedral. To work out the identification criteria for them is the aim of the following paper.

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* See deposition footnote.

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Chlorite Polytypism. II. Classification and X-ray Identification of Trioctahedral Polytypes

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Abstract

Classification and identification criteria are presented for tri-trioctahedral chlorite polytypes. According to the diffractions hkl ($k = 3n$) and $0kl$ ($k \neq 3n$) they can be categorized into four subfamilies and eight MDO (maximum degree of order) groups, respectively. Each subfamily has its characteristic superposition structure, each MDO group its characteristic YZ projection — geometrical properties of these can be derived from the fully descriptive polytype symbols. Identification diagrams for single-crystal specimens (taking into account also the variability in the chemical composition) are given for one- and two-layer polytypes. Characteristic properties of powder diffraction patterns useful for identification purposes are also given.

1. Introduction

Identification of chlorite polytypes is complicated by the variability in their chemical composition, which has to be taken into consideration, in contrast to vermiculites (Weiss & Ďurovič, 1980) whose composition lies within narrow limits. Accordingly, the aim of this

paper is to respect this fact when working out identification criteria for chlorite polytypes. The basis for this task are lists of MDO polytypes derived in part I (Ďurovič, Dornberger-Schiff & Weiss, 1983). Since the majority of natural chlorite specimens identified so far are trioctahedral, only these will be considered in the following.

2. Geometrical background for classification of chlorite polytypes

It is commonly known that diffraction patterns of chlorites consist of two sets of diffractions. Those with $k = 3n$ (orthogonal indexing) are always sharp, whereas those with $k \neq 3n$ are sharp only for periodic polytypes. According to Brown & Bailey (1962) the diffractions with $k = 3n$ and $h \neq 3n$ are characteristic for each of their four assemblages Ia, Ib, IIa, IIb, while the diffractions with $k \neq 3n$ are characteristic for individual polytypes.

Systematic calculations of diffraction patterns for maximum degree of order (MDO) polytypes of Mg-vermiculite (Weiss & Ďurovič, 1980) revealed that according to the diffractions with $k = 3n$, in particular