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ON THE NUMBER OF DISTINCT POLYTYPES OF MICA AND SiC WITH A PRIME LAYER-NUMBER*

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Abstract

To obtain formulae that give the numbers of distinct polytypes of mica and SiC when the layer-number p is a prime, a so-called symbol-differentiating operation is used to derive different layer-stacking symbols that represent one and the same polytype. If a layer-stacking symbol ω_2 is produced as a result of application of a symbol-differentiating operation ϵ to a symbol ω_1, ω_1 and ω_2 are said to be equivalent to each other, and the set Ω of all possible symbols is partitioned into disjoint equivalence classes. A one-to-one correspondence exists between the set of different polytypes theoretically possible and the set of equivalence classes in Ω. For mica polytypes with a prime layer-number, the equivalence classes in Ω were grouped into six families, and the number of classes within each of these families was determined. The number of distinct mica polytypes was thus derived when the stacking operation lement entre couches contigües est une rotation of 60° (senary) or 120° (ternary), respectively. Corresponding numbers are obtained for SiC polytypes, and the formulae are derived for SiC polytypes of any layer-number.

Sommaire

Dans le but de trouver des formules donnant le nombre des polytypes distincts de mica, ou de SiC, lorsque le nombre p de couches est premier, on emploie une opération dite "différenciatrice de symboles" pour établir des symboles d'empilement de couches différents qui représentent un seul et même polytype. Soit ω_2 le symbole d'empilement que l'on obtient en appliquant l'opération différenciatrice ϵ au symbole ω_1, on dit alors que ω_1 et ω_2 sont équivalents l'un à l'autre, et l'ensemble Ω de tous les symboles possibles est partagé en classes d'équivalence disjointes. Une correspondance bi-univoque relie l'ensemble des polytypes théoriquement possibles à l'ensemble des classes d'équivalence de Ω. Dans les cas des polytypes de mica où p est nombre premier, les classes d'équivalence de Ω se groupent en six familles, et nous avons déterminé le nombre de classes que contient chaque famille. Ainsi s'établit le nombre des polytypes distincts de mica lorsque l'opération d'empilement entre couches contigües est une rotation d'un multiple de 60° (sénaire) ou de 120° (ternaire). Quant aux polytypes de SiC, nous en avons déterminé les nombres correspondants et établi les formules pour n'importe quel nombre p.

(Traduit par la Rédaction)

Introduction

A complex structure very commonly consists of substructures. Such a structure may be called a hierarchic structure and is in most cases responsible for polytypism, twinning or domain texture. A study of mathematical principles for representation of complex structures such as the polytypes of SiC, ZnS and mica has been considered to be one of the important problems of modern crystallography. In these polytypes, unit layers are stacked upon each other in one

*Dedicated to Professor J. D. H. Donnay on the occasion of his 75th birthday.

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of a few different orientations, and their sequence repeats after a certain number of layers. These phenomena may be attributed to the spiral-growth mechanism (Frank 1951). Several systems of notations to describe these polytypes have been proposed: layer-orientation symbol (Zvyagin 1960), vector-stacking symbol (Ross *et al.* 1966) and layer-position symbol (Takeda & Sadanaga 1969) for mica, and *ABC* sequence, Zhdanov symbol and *h–k* notation for SiC (Verma & Krishna 1966).

A practical method of enumerating the possible sequences for *N*-layer mica polytypes was established by Ross *et al.* (1966). Takeda (1971) examined the algebraic properties of the vector-stacking symbol and introduced three operations, each of which produces a certain change of symbol while leaving the structure of the polytype invariant. By applying these operations he enumerated by computer the possible mica polytypes up to $N = 10$ for those with 0° and ±120° interlayer rotations (in the ternary representation) and up to $N = 8$ for the general case (senary). Enumeration of SiC polytypes has been carried out by computer up to $N = 26$ (Tokonami & Hosoya 1966), and the sequences and symmetries of stacked closest-packed layers are tabulated for *N* up to 12 in International Tables for X-ray Crystallography, Vol. II (1959).

Because the method adopted in these enumerations mentioned above is a kind of computer simulation, a theory capable of predicting the number of possible polytypes for a given layer-number has been much desired, in the hope that such a theory will reveal the mathematical principle governing the structures of polytypes, thus promoting a better understanding of the physical mechanism of their formation.

As a result of our recent collaboration, we have succeeded in deriving five kinds of formulae that meet our objectives: the first gives the number of mica polytypes in the senary representation and with a prime layer-number, the second the number of mica polytypes (ternary) with a prime layer-number, the third the number of mica polytypes (senary) with a layer-number relatively prime (Birkhoff & MacLane 1965) to 6, the fourth the number of SiC polytypes with a prime layer-number, and the fifth the number of SiC polytypes of any layer-number. However, because the example where *N* is a prime number is best suited for an intellible demonstration of the relation between different polytypes and the mathematical structure of the set of polytype symbols, we will expound, in this paper, the theory of enumerating mica and SiC polytypes with a prime layer-number.

Polytype Symbol and Symbol-Differentiating Operations

In order to present the system of symbols and operations for deriving and describing our theory, let us first give a brief review of the vector-stacking symbol (Ross *et al.* 1966) and Takeda's (1971) method of enumerating mica polytypes. In the following discussion we assume that *p*, the number of layers constituting a unit cell of a mica or SiC polytype under consideration, is always a prime.

The unit layer of the mica polytype is usually so chosen as to coincide with the unit slab in one-layer mica 1*M* and possessing the symmetry $1C12/m$ (A. Niggli's notation for the diperiodic groups). The structure of a mica polytype is then considered as a stacking of the unit layers with rotations by 0°, ±60°, ±120° or 180° between pairs of adjacent layers. Therefore the polytype symbol ω expressing the stacking sequence of layers in a *p*-layer mica has been given by a series of *p* numbers as

$$\omega = (a_1a_2 \ldots a_j \ldots a_{p-1}a_p), \tag{1}$$

where the *j*th number a_j refers to the angle of the relative rotation between the *j*th and the $(j+1)$th layer, the first layer being that in which the origin of the polytype structure is taken. The simplest expression will be to give a_j one of the values of 0, 1, 2, 3, 4 and 5 corresponding to rotations by 0°, 60°, 120°, 180°, 240° and 300°, respectively. Because of the periodicity along the direction of the layer stacking, the condition

$$\sum_{j=1}^{p} a_j \equiv 0 \pmod 6 \tag{2}$$

is imposed upon every *p*-layer polytype.

It will then be obvious that every symbol ω of such *p* numbers that satisfy (2) represents a possible mica polytype of *p* layers; in order to enumerate all possible *p*-layer polytypes, all ωs conformable to (2) must be generated. However the total number of different ωs thus generated will not coincide with but exceed the total number of different polytypes theoretically possible, because two or more symbols apparently different from each other may represent one and the same polytype, whereas one symbol cannot express more than one polytype. The truth of the second half of this statement will be obvious from the fact that a symbol determines one and only one type of sequence, thus only one polytype, and the truth of the first half will be demonstrated below with the aid of symbol-differentiating operations.

By symbol-differentiating we mean such an operation that preserves the layer sequence of a mica polytype under consideration but transforms its symbol ω to ω' which is not congruent with ω. The simplest of the symbol-differentiating operations will be derived when the fact that the origin can be taken in any arbitrarily chosen layer within the polytype is taken into account. If the choice of the origin in layer A_i is specified as different from the choice of it in A_j when $A_i \neq A_j$, every different choice of the origin in a mica polytype will create a unique symbol unless all the numbers in the symbol are the same. When an operation that shifts the origin by one layer is denoted by ρ,

$$(a_1a_2 \ldots a_{p-1}a_p)\rho = (a_2 \ldots a_{p-1}a_pa_1) \qquad (3)$$

and

$$(a_1a_2 \ldots a_ia_{i+1} \ldots a_{p-1}a_p)\rho^i = (a_{i+1} \ldots a_{p-1}a_pa_1a_2 \ldots a_i) \qquad (4)$$

hold. If all a_js in (3) are not equal to each other, each of ρ^is $(i = 1, \ldots, p\text{-}1)$ will be a symbol-differentiating operation, because p is a prime and accordingly no symbol can have an internal period. The operations ρi are the only symbol-differentiating operations that can be derived from a polytype in a fixed position, because the possibility of a change of symbol then lies only in a shift of the origin.

Next we must give the polytype a motion and see if we can derive more kinds of symbol-differentiating operations. Suppose first that the motion is of the first kind. Because the symbol is a string of numbers in one direction, the only motion of the first kind and representative of those producing a change of symbol is a rotation by 180° around an axis parallel to the layers. If this rotation is denoted by τ, it will operate on the symbol as

$$(a_1a_2 \ldots a_{p-1}a_p)\tau = (a_pa_{p-1} \ldots a_2a_1) \qquad (5)$$

and therefore it is a symbol-differentiating operation for those symbols that are not reflection-symmetrical across a plane perpendicular to it. Suppose next that the motion is of the second kind. Because of the symmetry $1C12/m$ of the unit layer, the motion of the second kind and representative of those compatible with the structures of some mica polytypes is a reflection across a plane perpendicular to the layers, but this motion is not symbol-differentiating because the structures of these polytypes and their symbols as well are then symmetrical with respect to this reflection. Then the fact must be taken into account that two polytypes enantiomorphic with each other are looked upon as having one and the same layer sequence; if a mica polytype is not reflection-symmetrical, the operation ϵ that creates its enantiomorph will be a symbol-differentiating operation. If we define $\bar{a}_j = 6\text{-}a_j$ when $a_j \neq 0$, and $\bar{a}_j = a_j$ when $a_j = 0$, we shall have a relation

$$(a_1a_2 \ldots a_{p-1}a_p)\epsilon = (\bar{a}_1\bar{a}_2 \ldots \bar{a}_{p-1}\bar{a}_p), \qquad (6)$$

where the structure represented by the symbol on the right side is enantiomorphic with that given by the symbol on the left side. Three kinds of symbol-differentiating operations, ρ, τ and ϵ, have thus been derived for mica polytypes, and from the ways they were derived, it is obvious that they and their combinations exhaust all the operations required.

Next let us turn to SiC polytypes. As no adjacent layers in the SiC structure can take the same letter in the *ABC* sequence, two arbitrarily-selected adjacent layers will be expressed by *AB*. The layer to follow *AB* is then either *C* or *A*, providing sequences *ABC* or *ABA*. Depending on the third layer upon AB, C or A, the mode of its stacking upon *AB* will here be denoted by 0 or 1, respectively. The polytype symbol of SiC will thus be a string of p numbers, each of which is either 0 or 1. We will then introduce an experimentally obtained rule and assume that SiC polytypes formed at high temperatures contain no 1 in their Zhdanov symbols, which is equivalent to saying that no two 1s can be adjacent to each other in our polytype symbol. Though the operations ρ and τ apply also to the present case, ϵ leaves the symbol of SiC invariant as obvious from its definition; accordingly, it is not symbol-differentiating.

Enumeration of p-Layer Mica Polytypes

Our aim is to enumerate p-layer polytypes of mica from the set Ω of polytype symbols generated under the assumption that p is a prime, and under the condition (2). This problem can be solved through a series of procedures purely mathematical in nature. However, because the complete presentation of proofs of all the lemmas and theorems necessary for this purpose would require too lengthy a description, we shall confine ourselves chiefly to the demonstration of those of special importance, thereby concentrating our main effort into a systematic description of our logical sequence. The theory will first be developed for mica polytypes, and as a variation of it SiC polytypes will be dealt with later.

Let p be a prime larger than 4, and consider a set Ω of polytype symbols in the form of (1), for which (2) holds. Such operations ρ, τ and ϵ are then defined on the elements (polytype symbols) of Ω as expressed by (3), (5) and (6) respectively. If applications, in succession and in finite repetitions if required, of ρ, τ and ϵ to one element ω_1 of Ω produce ω_2, ω_2 is said to be equivalent to ω_1, $\omega_2 \sim \omega_1$. Then the relation expressed by $\sim$ is an equivalence relation in Ω, by which Ω is partitioned into disjoint equivalence classes Γ_i s, for example such as

$$\Omega = \Gamma_1 \mathrm{U} \Gamma_2 \mathrm{U} \ldots\ldots \mathrm{U} \Gamma_n, \tag{7}$$

and all such ωs that are equivalent to ω_i constitute the ith class Γ_i. Then because the operations ρ, τ and ϵ are symbol-differentiating operations, all such ωs that are different from but equivalent to each other represent one and the same polytype on the one hand and belong to the same equivalence class on the other. This means that a 1:1 correspondence can be established between the set of different polytypes and the set of equivalence classes given in Ω by the equivalence relation due to ρ, τ, ϵ and their combinations. Therefore the problem of enumerating different polytypes can be identified as that of seeking the total number of equivalence classes in (7). When Γ_i is looked upon as a subset of Ω, the number of different elements in Γ_i is called the length of Γ_i and is denoted by $|\Gamma_i|$.

In the previous section ρ, τ and ϵ were defined as symbol-differentiating operations, but they should now be redefined as operations given by the defining relations (3), (5) and (6) respectively. Therefore the identity 1 defined by

$$\rho^p = \tau^2 = \epsilon^2 = 1, \tag{8}$$

should now be looked upon as a legitimate member of the operations under consideration. Also from (3), (4), (5) and (6) the following relations will be immediately recognized:

$$\rho\tau = \tau\rho^{-1} \tag{9}$$

and

$$\rho\epsilon = \epsilon\rho, \ \tau\epsilon = \epsilon\tau. \tag{10}$$

From (8), (9) and (10) it will be directly proved that every product of finite numbers of ρ, τ and ϵ in an arbitrary order coincides with one of the following $4p$ operations:

$$\begin{array}{l} 1, \rho, \rho^2, \ldots\ldots, \rho^{p-1}, \\ \tau, \rho\tau, \rho^2\tau, \ldots\ldots, \rho^{p-1}\tau, \\ \epsilon, \rho\epsilon, \rho^2\epsilon, \ldots\ldots, \rho^{p-1}\epsilon, \\ \tau\epsilon, \rho\tau\epsilon, \rho^2\tau\epsilon, \ldots\ldots, \rho^{p-1}\tau\epsilon. \end{array} \tag{11}$$

Thus, from now on, we shall deal with only these $4p$ operations, denote the sets of operations in the first, second, third and fourth rows in (11) by P, T, E and TE respectively, and express the set of symbols $\{\omega, \omega\rho, \ldots, \omega\rho^{p-1}\}$ as ωP and in a similar fashion for the other rows.

The enumeration of equivalence classes will then be carried out in two steps: (I) derivation of families of classes according to combinations of defining relations, and (II) determination of the number of classes in each of the families. Before proceeding to the description of these steps, it will be appropriate to refer to the following two lemmas because the first lemma is very often used in (I) and the second proves that A to F in (I) exhaust all the possible kinds of family.

Lemma 1: When $\omega\rho \neq \omega$, all elements in ωP are different from each other.

Proof: If $\omega\rho^i = \omega\rho^j$ $(0 \leq i < j \leq p-1)$, $\omega\rho^{j-i} = \omega$ $(1 < j-i \leq p-1)$. As p and $j-i$ are relatively prime, there exist such integers m and l that satisfy $m(j-i) + lp = 1$. Then $\omega = \omega\rho^{j-i} = \omega(\rho^{j-i})^m = \omega\rho^{(j-i)m} = \omega\rho^{1-lp} = \omega\rho$, contrary to the assumption that $\omega\rho \neq \omega$. Q.E.D.

Lemma 2: Among three operations τ, ϵ and $\tau\epsilon$, if two which are arbitrarily chosen leave ω invariant, the remaining one also leaves ω invariant. If only one of the two arbitrarily chosen operations leaves ω invariant, it is the only one of the three operations that leaves ω invariant.

Proof: When $\omega\tau = \omega$ and $\omega\tau\epsilon = \omega$, $\omega\tau\epsilon = \omega\epsilon = \omega$, *etc.*, and when $\omega\epsilon = \omega$ but $\omega\tau\epsilon \neq \omega$, $\omega\tau\epsilon = \omega\epsilon\tau = \omega\tau \neq \omega$, *etc.* Q.E.D.

(I) Derivation of Families of Classes According to Combinations of Defining Relations

(*A*) $\omega\rho = \omega$, $\omega\tau = \omega$, *and* $\omega\epsilon = \omega$

From $\omega\rho = \omega$, $a_1 = a_2$, $a_2 = a_3, \ldots, a_p = a_1$. From (2), $\sum_{j=1}^{p} a_j = pa_1 \equiv 0 \pmod 6$. Because p is a prime and $a_1 \leq 5$ by assumption, it is concluded that $a_1 = 0$ and $\omega = (00\ldots00)$. Conversely it is obvious that $\omega = (00\ldots00)$ is left invariant by every operation in (11). Hence under (*A*), $\{\omega = (00\ldots00)\}$ constitutes a class Γ_A of length 1. (Because this sequence represents the one-layer polytype, it should be eliminated from the total numbers of the p-layer polytypes).

(*B*) $\omega\rho \neq \omega$, $\omega\tau = \omega$, *and* $\omega\epsilon = \omega$

The relations $\omega\tau = \omega$ and $\omega\epsilon = \omega$ make each of ωT, ωE and ωTE coincide with ωP. Obviously ρ leaves ωP invariant. When $\omega\tau = \omega$, τ leaves ωP invariant; $\omega\rho^i\tau = \omega\tau\rho^{-i} = \omega\rho^{-i} \in \omega P$. The same applies to ϵ when $\omega\epsilon = \omega$ and to $\tau\epsilon$ when $\omega\tau = \omega$ and $\omega\epsilon = \omega$. Hence ωP is left invariant by every operation in (11) and is an equivalence class Γ_B under (*B*). Then as $\omega\rho \neq$

ω, all elements of ωP are different from each other by Lemma 1. Hence $|\Gamma_B| = p$.

(*C*) $\omega\rho \neq \omega$, $\omega\tau = \omega$, and $\omega\epsilon \neq \omega$

The relation $\omega\tau = \omega$ makes ωT and ωTE coincide with ωP and ωE respectively. Obviously ρ leaves both ωP and ωE invariant. It may then be easily proved, as before, that when $\omega\tau = \omega$, τ leaves both ωP and ωE invariant. On the other hand ϵ maps ωP onto ωE and *vice versa* and the same applies to $\tau\epsilon$ when $\omega\tau = \omega$. Therefore $\omega P \cup \omega E$ is left invariant by every operation in (11) and is an equivalence class Γ_C under (*C*). As $\omega\rho \neq \omega$, all elements in ωP are different from each other. If $\omega\rho^i\epsilon = \omega\rho^j$ $(1 \leq j < i < p-1)$, $\omega = \omega\rho^{i-j}\epsilon = \omega(\rho^{i-j}\epsilon)^p = \omega(\rho^{i-j})^p\epsilon^p = \omega\epsilon$, contrary to $\omega\epsilon \neq \omega$ in (*C*). (It should be noted that ρ and ϵ are commutative.) Thus all elements in $\omega P \cup \omega E$ are different from each other and $|\Gamma_C| = 2p$.

(*D*) $\omega\rho \neq \omega$, $\omega\tau\epsilon = \omega$, *and* $\omega\epsilon \neq \omega$

As $\omega\tau\epsilon = \omega$ makes ωTE and ωT coincide with ωP and ωE respectively, exactly the same argument holds as in (*C*), which leads to the conclusion that Γ_D is $\omega P \cup \omega E$ and $|\Gamma_D| = 2p$. It is to be noted that Γ_C and Γ_D can have no element in common; by Lemma 2, $\omega\tau = \omega$ and $\omega\tau\epsilon \neq \omega$ in (*C*) whereas $\omega\tau \neq \omega$ and $\omega\tau\epsilon = \omega$ in (*D*), and obviously no element can satisfy these two sets of mutually inconsistent conditions.

(*E*) $\omega\rho \neq \omega$, $\omega\tau \neq \omega$ *for every element of* Γ_E, *and* $\omega\epsilon = \omega$

The relation $\omega\epsilon = \omega$ makes ωE and ωTE coincide with ωP and ωT respectively. Then ρ leaves each of ωP and ωT invariant, the same applies to ϵ when $\omega\epsilon = \omega$, but τ maps ωP onto ωT and *vice versa*, and the same applies to $\tau\epsilon$ when $\omega\epsilon = \omega$. Therefore $\omega P \cup \omega T$ is left invariant by every operation in (11) and is an equivalence class Γ_E under (*E*). As $\omega\rho \neq \omega$, all elements in ωP are different from each other. If $\omega\rho^i = \omega\rho^j\tau$, $\omega\rho^{i+j}\tau = \omega$. Take such integers m and l that satisfy $2m + lp = 1$, and put $\omega' = \omega\rho^{(i+j)m}$. Then $\omega' \in \omega P \subset \Gamma_E$; $\omega'\tau = \omega\rho^{(i+j)m}\tau = \omega\rho^{2(i+j)m}\tau\rho^{(i+j)m} = \omega\rho^{(i+j)(1-lp)}\tau\rho^{(i+j)m} = \omega\rho^{(i+j)}\tau\rho^{(i+j)m} = \omega\rho^{(i+j)m} = \omega'$, contrary to $\omega\tau \neq \omega$ for every element of Γ_E. (It should be noted that as ρ and τ are not commutative, the above part of the proof cannot be carried out in the same way as for the corresponding part in (*C*) but calls for such an element ω' that is equal to $\omega\rho^{(i+j)m}$ and satisfies $\omega'\tau \neq \omega'$.) Thus all elements in $\omega P \cup \omega T$ are different from each other and $|\Gamma_E| = 2p$.

(*F*) $\omega\rho \neq \omega$, $\omega\tau \neq \omega$ *and* $\omega\tau\epsilon \neq \omega$ *for every element of* Γ_F, *and* $\omega\epsilon \neq \omega$

Operation ρ leaves invariant each of ωP, ωT, ωE and ωTE. Operation τ maps ωP onto ωT, ωT onto ωP, ωE onto ωTE, and ωTE onto ωE. Operation ϵ maps ωP onto ωE, ωT onto ωTE, ωE onto ωP, and ωTE onto ωT. Operation $\tau\epsilon$ maps ωP onto ωTE, ωT onto ωE, ωE onto ωT, and ωTE onto ωP. Hence only $\omega P \cup \omega T \cup \omega E \cup \omega TE$ will in this case be left invariant by every operation in (11) and is an equivalence class Γ_F under (*F*). Relations $\omega\rho \neq \omega$ and $\omega\epsilon \neq \omega$ assure as proved in (*C*) that all elements in $\omega P \cup \omega E$ are different from each other. Relations $\omega\rho \neq \omega$ and $\omega\tau \neq \omega$ assure as proved in (*E*) that all elements in $\omega P \cup \omega T$ are different from each other. Likewise, relations $\omega\rho \neq \omega$ and $\omega\tau\epsilon \neq \omega$ will be easily proved to assure that all elements in $\omega P \cup \omega TE$ are different from each other. When ωP and ωT are operated upon by ϵ, ωE and ωTE will be produced. Therefore all elements in $\omega E \cup \omega TE$ are different from each other. When ωP and ωE are operated upon by τ, ωT and ωTE will be produced. Therefore all elements in $\omega T \cup \omega TE$ are different from each other. When ωP and ωTE are operated upon by $\tau\epsilon$, ωE and ωT will be produced. Therefore all elements in $\omega E \cup \omega T$ are different from each other. Hence all the $4p$ elements in Γ_F are different from each other and $|\Gamma_F| = 4p$. Thus we have the following theorem.

Theorem 1: Equivalence classes in Ω produced by operations in (11) are classified into six families as follows: Family *A* defined by ($\omega\rho = \omega$, $\omega\tau = \omega$, and $\omega\epsilon = \omega$) and consisting of a class of length 1; Family *B* defined by ($\omega\rho \neq \omega$, $\omega\tau = \omega$, and $\omega\epsilon = \omega$) and consisting of classes of length p; Family *C* defined by ($\omega\rho \neq \omega$, $\omega\tau = \omega$, and $\omega\epsilon \neq \omega$) and consisting of classes of length $2p$; Family *D* defined by ($\omega\rho \neq \omega$, $\omega\tau\epsilon = \omega$, and $\omega\epsilon \neq \omega$) and consisting of classes of length $2p$; Family *E* defined by ($\omega\rho \neq \omega$, $\omega\tau \neq \omega$ for every element of Γ_E, and $\omega\epsilon = \omega$) and consisting of classes of length $2p$, and Family *F* defined by ($\omega\rho \neq \omega$, $\omega\tau \neq \omega$ and $\omega\tau\epsilon \neq \omega$ for every element of Γ_F, and $\omega\epsilon \neq \omega$) and consisting of classes of length $4p$.

(II) Determination of the Number of Classes in Each of the Families

Family A

Family *A* contains only one class $\{\omega = (00\ldots00)\}$. Hence, its class number n_A is equal to 1.

Family B

In order to count the number n_B of classes in this family, the fact must be utilized that every class in this family contains one and only one such element ω that satisfies $\omega\tau = \omega$ and $\omega\epsilon = \omega$, which means that n_B is equal to the total number of elements satisfying these two conditions except $\omega = (00\ldots00)$. In fact if $\omega\rho^i\tau = \omega\rho^i$ $(1 < i \leq p-1)$, $\omega\rho^{2i}\tau = \omega = \omega\tau$. Thus $\omega\rho^{2i} = \omega$. Take such integers m and l that satisfy $m(2i) + lp = 1$. Then $\omega = \omega\rho^{2i} = \omega(\rho^{2i})^m = \omega\rho^{1-lp} = \omega\rho$, contrary to (B). Q.E.D. Let us therefore count the number of these elements. Because $\omega\epsilon = \omega$, a_j in ω is either 0 or 3, and from $\omega\tau = \omega$ the equalities hold: $a_1 = a_p$, $a_2 = a_{p-1}, \ldots, a_{(p-1)/2} = a_{(p+3)/2}$. When each of $a_1, a_2, \ldots, a_{(p-1)/2}$ is given the value of either 0 or 3, the remaining central number $a_{(p+1)/2}$ is determined by (2). Thus the number of elements that satisfy both $\omega\tau = \omega$ and $\omega\epsilon = \omega$ is $2^{(p-1)/2}$, because ω is determined by the first $(p-1)/2$ a_js and each of such a_js can have the alternative of 0 or 3. As this number includes n_A $(= 1)$, n_B is given by $n_B = 2^{(p-1)/2} - 1$.

Family C

In each of the equivalence classes in this family, those elements that can satisfy both $\omega\tau = \omega$ and $\omega\epsilon \neq \omega$ are only two, ω and $\omega\epsilon$. In fact from (C) in (I) the equivalence class containing ω is expressed as $\omega P \cup \omega E$. If $\omega\rho^i$ $(1 < i \leq p-1)$ in ωP satisfies $\omega\tau = \omega$, then $\omega\rho^i\tau = \omega\rho^i$, but this leads to $\omega\rho = \omega$ as proved in the previous case of family B. However, as $\omega\rho \neq \omega$ in (C), it is concluded that $\omega\rho^i$ $(1 < i \leq p-1)$ cannot satisfy $\omega\tau = \omega$. As $\omega\rho^i\epsilon\tau = \omega\rho^i\tau\epsilon$, it will be immediately concluded that $\omega\rho^i\epsilon$ $(1 < i \leq p-1)$ in ωE cannot satisfy $\omega\tau = \omega$. On the other hand, ω satisfies $\omega\tau = \omega$ by assumption, and because $\omega\epsilon\tau = \omega\tau\epsilon = \omega\epsilon$, $\omega\epsilon$ also satisfies $\omega\tau = \omega$. Q.E.D. Therefore the number n_C of equivalence classes in this family is equal to half the number of such elements that satisfy both $\omega\tau = \omega$ and $\omega\epsilon \neq \omega$. Then the number of elements that satisfy $\omega\tau = \omega$ is $6^{(p-1)/2}$ because ω is determined by its first $(p-1)/2$ a_js as in the previous case of family B and each of such a_j s can have choice among six numbers, 0, 1, 2, 3, 4 and 5. Then the set of these $6^{(p-1)/2}$ elements includes n_B elements of family B and one element of family A. Hence the total number of such elements that satisfy both $\omega\tau = \omega$ and $\omega\epsilon \neq \omega$ is $6^{(p-1)/2} - n_B - 1 = 6^{(p-1)/2} - 2^{(p-1)/2}$. As n_C is half this number, $n_C = (\frac{1}{2})\{6^{(p-1)/2} - 2^{(p-1)/2}\}$.

Family D

Exactly the same argument holds in this case as in the previous case of family C when $\omega\tau = \omega$ is replaced by $\omega\tau\epsilon = \omega$, and the number n_D of equivalence classes in this family is given by $n_D = (\frac{1}{2})\{6^{(p-1)/2} - 2^{(p-1)/2}\}$.

Family E

Let the set of all elements that satisfy $\omega\epsilon = \omega$ be Ω'. Then in $\omega = (a_1a_2\ldots a_j\ldots a_{p-1}a_p) \in \Omega'$, each of a_js is either 0 or 3. Hence, the total number of elements in Ω' is 2^{p-1} because one of the p numbers $a_1, a_2, \ldots, a_p$ in ω is determined from the rest by (2), and each of the remaining $p-1$ numbers can have the alternative of 0 or 3. All the elements of classes in families A and B are contained in Ω' because (A) and (B) have $\omega\epsilon = \omega$ in common with Ω'. Hence, the number of those elements in Ω' that are contained in classes of family E is $2^{p-1} - p \times n_B - 1$, and because $2p$ of these elements form a class, the number n_E of equivalence classes in family E is given by $n_E = (\frac{1}{2}p)\{2^{p-1} - p \times n_B - 1\} = (\frac{1}{2}p)\{2^{p-1} - p \times 2^{(p-1)/2} + p - 1\}$.

Family F

The total number of elements in Ω is 6^{p-1} because one of the p numbers in ω is determined from the rest by (2) and each of the remaining $p-1$ numbers can have choice among six numbers, 0, 1, 2, 3, 4 and 5. As all the elements in classes of families A, B, C, D, E and F constitute Ω and because $|\Gamma_F| = 4p$, the number n_F of classes in family F is given by $n_F = (\frac{1}{4}p)\{6^{p-1} - 2p(n_E + n_D + n_C) - p \times n_B - n_A\} = (\frac{1}{4}p)[\{6^{p-1} - 2^{p-1}\} - 2p\{6^{(p-1)/2} - 2^{(p-1)/2}\}]$.

We have thus completed all the procedures of (*II*) and now reached the theorem that gives the total number of equivalence classes in Ω:

Theorem 2: The total number $n_S(p)$ of equivalence classes in Ω is given by $n_S(p) = n_A + n_B + n_C + n_D + n_E + n_F$, where $n_A = 1$, $n_B = 2^{(p-1)/2} - 1$, $n_C = n_D = (\frac{1}{2})\{6^{(p-1)/2} - 2^{(p-1)/2}\}$, $n_E = (\frac{1}{2}p)\{2^{p-1} - p \times 2^{(p-1)/2} + p - 1\}$ and $n_F = (\frac{1}{4}p)[\{6^{p-1} - 2^{p-1}\} - 2p\{6^{(p-1)/2} - 2^{(p-1)/2}\}]$. Hence the total number of polytypes

$$\begin{aligned} n'_S(p) &= n_S(p) - n_A \\ &= (\tfrac{1}{4}p)\{6^{p-1} + (2p) \times 6^{(p-1)/2} \\ &\quad + 2^{p-1} - 2p - 2\}. \end{aligned} \tag{12}$$

Note here that p is greater than 4.

To derive the above theorem, it was assumed that any two adjacent layers in mica polytypes are stacked together with a relative rotation of one of the multiples of 60°, which is a digit in the senary representation. However, in natural micas the rotation is usually by one of 0°, 120°, and 240°. For these, therefore, it should be assumed that a_js in ω can take the values of 0, 2 and 4, by which the stacking sequence can be expressed by a number in the ternary representation. For this case those procedures in steps (I) and (II) will be considerably simplified because the relation $\omega\epsilon = \omega$ cannot hold in this

TABLE 1. NUMBERS OF DISTINCT MICA POLYTYPES FOR GIVEN LAYER NUMBERS (N) IN THE TERNARY REPRESENTATION (BASE 3)

1	1
2	1
3	2
4	4
5	8
6	18
7	39
8	94
9	222
10	572
11	1 463
12	3 934
13	10 584
14	29 211
15	80 808
16	226 430
17	636 320
18	1 800 318
19	5 107 479
20	?
23	341 187 047
29	197 216 119 544
31	1 660 419 530 055
37	1 014 153 134 906 184
41	74 132 108 201 056 400
43	636 156 918 785 481 015
47	47 143 287 917 563 616 519
53	30 476 801 365 547 693 758 184
59	19 958 172 445 992 979 743 434 999
61	Number of 27 figures

TABLE 2. NUMBERS OF DISTINCT MICA POLYTYPES FOR GIVEN LAYER NUMBERS (N) IN THE SENARY REPRESENTATION (BASE 6)

1	1
2	3
3	6
4	26
5	83
6	402
7	1 776
8	9 212
9	47 311
10	254 721
11	1 378 142
12	7 574 768
13	41 884 605
14	?
15	?
16	?
17	41 487 751 175
18	?
19	1 336 320 261 618
20	?
23	1 430 670 875 381 804
29	52 939 157 060 432 805 605
31	1 782 854 191 531 337 908 752
37	69 692 059 449 311 155 754 627 625
41	81 509 113 041 731 913 865 429 529 075
43	2 797 847 694 176 606 471 071 811 477 694
47	3 317 413 963 852 624 100 552 130 498 223 256
53	137 255 311 267 601 444 345 446 666 542 784 075 505
59	5 752 551 551 399 394 713 921 759 959 900 326 815 503 238
61	Number of 45 figures

type of sequence. The final result corresponding to this case of the ternary representation is given by

$$n_T^1(p) = (\tfrac{1}{4}p)\,\{3^{p-1} + (2p) \times 3^{(p-1)/2} - 2p - 1\}, \tag{13}$$

with p greater than 4.

In Tables 1 and 2, numbers of distinct mica polytypes are listed for given layer-numbers. In these tables the numbers of polytypes in the ternary and senary representations, up to 19 and 12 layers, respectively, have been generated by computer, and those polytypes with a layer-number larger than the above are the results of calculations according to (12) and (13).

Enumeration of p-Layer SiC Polytypes

Let us express p-layer SiC polytypes also by (1), in which each of the numbers a_js is either 0 or 1 in the binary representation. Instead of (2), however, the condition of 'no two 1s being adjacent' holds for SiC. Equations (3), (4) and (5) apply to SiC but in (6) $\bar{a}_j = a_j$ for all j, that is, ϵ is not symbol-differentiating for the present case and should accordingly be deleted. Therefore, the equations in (10) are trivial and the latter two rows in (11) should be omitted. In (A) in (I), equation (2) must be replaced by the condition that $\omega \neq (11\ \ldots 11)$, $2H$ being excluded from the beginning. Because the cases of SiC can be interpreted as always satisfying $\omega\epsilon = \omega$, only families A, B and E will be present in Theorem 1.

After Theorem 1 we shall depart from the thread of argument given to mica polytypes and resort to a method much simpler than that described in the previous section. When the condition of 'no two 1s being adjacent' is imposed, the sequence of p numbers, each of which is either 0 or 1, is equivalent to the well-known sequence called a PM sequence (sequence of plus and minus signs) (Berman & Fryer 1972), which is a typical Fibonacci sequence. Let the total number of sequences with p numbers be $f(p)$; $f(p) = f(p-1) + f(p-2)$ holds. As $f(1) = 2$ and $f(2) = 3$, if the mth number in the standard Fibonacci sequence, 1, 1, 2, 3, 5, ...is denoted by $F(m)$, $f(p) = F(p+2)$. On the other hand because a_1 and a_p cannot be 1 at the same time, those sequences with $a_1 = 1$, $a_2 = 0$, $a_{p-1} = 0$ and $a_p = 1$ must be excluded from $f(p)$. The number of such sequences is $f(p-4) = F(p-2)$. Hence the total number $|\Omega|$ of possible sequences is

$|\Omega| = F(p+2) - F(p-2) = F(p+1) + F(p) - \{F(p) - F(p-1)\} = F(p+1) + F(p-1)$. The number $|\Omega|$ consists of elements of classes of family E, those of classes of family B, and $(00 \ldots 00)$ of family A. The number of classes whose elements are left invariant by operation τ, $\omega\tau = \omega$, is given by $f\left(\dfrac{p-1}{2}\right)$

$= F\left(\frac{p-1}{2} + 2\right) = F\left(\frac{p+3}{2}\right)$, because each of such classes is determined when $a_2, a_3, \ldots, a_{(p+1)/2}$ are given, both a_1 and a_p being necessarily zero. Hence, when the layer-number p is a prime larger than 2, the number $n_B(p)$ of possible SiC polytypes (always in a binary representation) will be given as

$$\begin{aligned} n_B(p) &= (\tfrac{1}{2}p)\,[F(p+1) + F(p-1) \\ &\quad - p\left\{F\left(\frac{p+3}{2}\right) - 1\right\} \\ &\quad - 1] + F\left(\frac{p+3}{2}\right) - 1 \\ &= (\tfrac{1}{2})\ \{F(p+1) + F(p-1) \\ &\quad + pF\left(\frac{p+3}{2}\right) - p - 1\}, \end{aligned} \tag{14}$$

neither $3C$ nor $2H$ being counted in it.

As far as SiC polytypes under the condition of 'no two 1s being adjacent' are concerned, we have lately succeeded in solving the problem of their enumeration for any layer-number. Denote the total number of N-layer SiC polytypes by $n_B(N)$. Then

$$n_B(N) = n_{N,N} + n_{N,2N}, \tag{15}$$

$$n_{N,N} = F\left(\frac{N+3}{2}\right) - \Sigma' n_{l,l}$$

when N is odd, and

$$= F\left(\frac{N+4}{2}\right) - \Sigma' n_{l,l}$$

when N is even, (16)

and

$$\begin{aligned} n_{N,2N} &= (1/2N)\,\{F(N+2) - F(N-2) \\ &\quad - N \times n_{N,N} - \Sigma'(l \times n_{l,l} \\ &\quad + 2l \times n_{l,2l})\}, \end{aligned} \tag{17}$$

where $n_{i,j}$ expresses the number of sequences with a period of i layers and having j distinct representations and starts with $n_{1,1} = 1$, $n_{1,2} = 0$, $n_{2,2} = 1$ and $n_{2,4} = 0$, $F(k)$ is the kth number of the Fibonacci sequence, 1, 1, 2, 3, 5, ... and Σ' means a summation over all such ls that divide N, i.e., l/N, with $l \neq N$.

In Table 3, numbers of distinct SiC polytypes for given layer-numbers are listed. The numbers of these polytypes were derived according to (14), (15), (16) and (17), and those with N up to 43 were also generated by computer and confirmed to coincide with the above results of calculations.

TABLE 3. NUMBERS OF DISTINCT SiC POLYTYPES FOR GIVEN LAYER-NUMBERS (N)

N		N	
1	1	31	49 352
2	1	32	77 337
3	1	33	120 694
4	1	34	189 540
5	2	35	296 847
6	2	36	466 930
7	4	37	733 362
8	5	38	1 155 355
9	7	39	1 818 562
10	10	40	2 868 918
11	15	41	4 524 080
12	20	42	7 145 780
13	30	43	11 285 526
14	43	44	17 845 317
15	60	45	28 220 044
16	91	46	44 668 419
17	132	47	70 716 649
18	197	48	112 038 102
19	290	49	177 548 408
20	440	50	281 533 868
21	651	51	446 544 706
22	1 005	53	1 124 865 050
23	1 509	59	18 130 295 895
24	2 328	61	45 908 862 054
25	3 542	67	750 014 690 474
26	5 504	71	4 851 043 787 277
27	8 433	73	12 352 235 686 604
28	13 151	79	204 817 314 892 976
29	20 318	83	1 336 183 557 337 451
30	31 759	89	22 360 422 113 307 176
		97	# of 18 figures
		101	# of 19 figures

Conclusion

The present mathematical treatment leads to the same numbers of distinct polytypes for layer-number N up to 5 in the senary case, and for N up to 7 in the ternary one, as in earlier, more direct treatments by Ross *et al.* (1966), and for N up to 8 (senary) and for N up to 10 (ternary), respectively, as in a computer simulation by Takeda (1971). The numbers of mica polytypes generated by computer with our improved program for the senary case agree with those given by our formulae for N up to 11 and for the ternary one for N up to 19.

In order to make practical application of these results, the complete set of a listing of the specific mica polytypes for $N = 5$ and 6 is available, at a nominal charge, from the Depository of Unpublished Data, CISTI, National Research Council of Canada, Ottawa, Canada, K1A 0S2.

We believe that the treatments leading to Theorem 2 in this paper will be successfully applied to all variations of polytype provided appropriate reinterpretations of the meaning of some of the quantities in this theory are introduced, if they are required at all.

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