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Fe-Li micas: a new approach to the substitution series

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ABSTRACT

Over the last 50 years many definitions of element substitution series have been proposed for describing natural Fe-Li micas with complex chemical compositions. In order to compare these definitions and ascertain their reliability, a new geometric frame composed of ideal mica points, mica joins (segments), substitution vectors and substitution planes is constructed to express the entire substitution system of ideal Fe-Al-Li micas. The frame is built in composition space with the coordinate system (0; Si, Al_{VI}, Al_{IV}, Fe^{2+}, Li, $\square_{VI}$, K), but has a 3-dimensional analogue, i.e. a visual image.

Using this frame, it has been proved that there are only five possible types of replacement for definitions of Fe-Li micas, and that all possible types have been suggested except the Al_{IV}-constant. Furthermore, it has been proved that the main point in different definitions is that the replacement of Li by Fe^{2+} needs to be balanced with Al_{IV}, Al_{VI}, or $\square_{VI}$. In order to solve this problem, a set of formulae determining spatial relations between geometric elements in the frame is suggested. With these formulae, the abstract frame is suggested to be a datum system used to ascertain quantitatively the reliability of definition of natural Fe-Li micas.

KEYWORDS: Fe-Li micas, substitution series, substitution vector, substitution plane.

Introduction

FE-LI micas are the typomorphic minerals of rare-metal granite, pegmatite, greisen etc. Rieder *et al.* (1999) calls the Fe-Li micas the 'zinnwaldite series'. Over the last 50 years, many authors have tried to define the Fe-Li mica series, and to clarify essential replacements within the series. None of the other mica series present so great a challenge as the Fe-Li mica series does in terms of interpretation of its crystallochemistry. We attempt to differentiate various 'Fe-Li mica series' and to lay a foundation for determining reliable relationships in terms of replacements, among natural Fe-Al-Li micas. For these purposes, a new geometric frame of Fe-Al-Li micas is constructed. The frame has two forms related to each other: one is abstract in space, with coordinate systems (0; Si, Al_{VI}, Al_{IV}, Fe^{2+}, Li, $\square_{VI}$, K) and the other is visual (Fig. 1*a*). To define a trustworthy replacement for the Fe-Li mica series is beyond the means of this paper, for it is necessary to determine the variation trend of natural Fe-Li micas (cf. Sun and Yu, in prep.).

Review

Fe-Li micas have been variously called the 'protolithionite-lepidolite series' (Winchell, 1927), the 'biotite-lepidolite series' (Ginzburg and Berkhin, 1953; Lapides *et al.*, 1977; Sun, 1988) and the 'siderophyllite-polylithionite series' (Foster, 1960*a*; Rieder, 1970; Stone *et al.*, 1988) with different replacements.

Ginzburg and Berkhin (1953) considered that the transition from biotite (having Al contents between 15 and 25% and R^{2+} between 85 and 75% in its octahedral sheet) to lepidolite was due to replacement (1):

$$2Mg = Li + Al_{VI} \quad (1)$$

Foster (1960*a*, Fig. 37) described Fe-Li micas in her triangle with apices Li, $R^{2+}{}_{VI}$, and $(R^{3+}+Ti)_{VI}$ as a band trending diagonally from

the area occupied by siderophyllite and aluminian lepidomelane towards the point representing polylithionite. She explained the band with equation (2). Since the ideal end-member is polylithionite (Pol, No. 2 in Table 1), ideal siderophyllite as a starting member of her series is formula (3) (Sid_0, No.11 in Table 1). Siderophyllite is also the Fe^{2+}-highest member of the Mg-Fe mica series (Foster, 1960*b*).

$$4n\mathrm{Li} + 2n\mathrm{Si} = 3n\mathrm{Fe}^{2+} + 2n\mathrm{Al}_{iv} \quad (2)$$

$$\mathrm{K_2Al_2Fe_3^{2+}\square_{VI}\,Si_6Al_2O_{20}(OH)_4} \quad (3)$$

Rieder (1970) and Stone *et al.* (1988) interpreted Fe-Li micas as generalized formulae (4) and (5) ($0 \leqslant X \leqslant 4$), respectively. According to their interpretations, Fe-Li micas are a solid-solution series from formula (6) (when $X = 4$ in formula (4) or No. 4 in Table 1) to Pol (when $X = 0$), which is caused by equation (7):

$$\mathrm{K_2Fe_X^{2+}Li_{4-X}(Al,\,Fe^{3+})_2Al_XSi_{8-X}O_{20}(OH,F)_4} \quad (4)$$

$$\mathrm{K_2\,R_X^{2+}Li_{4-X}\,R_2^{3+}Al_X\,Si_{8-X}O_{20}(OH,\,F)_4} \quad (5)$$

$$\mathrm{K_2Al_2Fe_4^{2+}Si_4Al_4O_{20}(OH)_4} \quad (6)$$

$$\mathrm{Fe^{2+} + Al_{IV} \rightarrow Li + Si} \quad (7)$$

Formula (6) is the end-member mineral called siderophyllite (e.g. Rieder *et al.*, 1999). Starting from annite, $\mathrm{K_2Fe_6Si_6\,Al_2O_{20}\,(OH)_4}$, formula (6) and Sid_4 (No. 15 in Table 1) are formed by Fe-Tschermak exchange, $2Si + 2Fe^{2+} \rightarrow 2Al_{IV} + 2Al_{VI}$, whereas varieties of siderophyllite Sid_0

TABLE 1. Definitions of ideal Fe-Al-Li micas used in this paper

No.	Ideal mineral	Abbreviation	Structural formula							Note
			K	Al_{VI}	Fe^{2+}	Li	$\square_{VI}$	Si	Al_{IV}	
End-member minerals										
1	Annite	Ann, A	2		6			6	2	
2	Polylithionite	Pol, P	2	2		4		8		
3	Trilithionite	Tri, T	2	3		3		6	2	
4	Siderophyllite*	Fe-Eas, E	2	2	4			4	4	*
5	Muscovite	Mus, M	2	4			2	6	2	
6	Aluminoceladonite	Al-Cel	2	2	2		2	8		
Other ideal minerals										
7	Zinnwaldite (Betehtin, 1950)	Zin, Z	2	2	2	2		6	2	Zin =Ann+2AT/3 =Fe-Eas+ZP
8	Protolithionite (Winchell, 1942)	Pro	2	1	4	1		6	2	Pro=Ann+AT/3 =Sid_4+ET/3
9	Varieties of protolithionite	Pro1, P_{r1}	2	2	5/2	1	1/2	6	2	Pro1=Pro+AM/4 =Sid_0+SZ/2
10	Al-lepidolite† Lapides *et al.* (1977)	Al-Lep	2	3		2	1	7	1	Al-Lep = Mus+MP/2
11	Varieties of	Sid_0	2	2	3		1	6	2	Sid_0 = Ann+AM/2
12	siderophyllite	Sid_1	2	4/3	4		2/3	6	2	Sid_1=Ann+AM/3
13		Sid_2	2	4/9	16/3		2/9	6	2	Sid_2=Ann+AM/9
14		Sid_3	2	2	10/3		2/3	16/3	8/3	Sid_3=Sid_0−ES/3 =Sid_4+AM/3
15		Sid_4	2	2/3	16/3			16/3	8/3	Sid_4=Ann+AE/3
16		A1	2	3/2	4		1/2	11/2	5/2	A1=Sid_1−ES_1/4
17		E1	2	5/3	23/6		1/2	16/3	8/3	E1=A1+AE/12
18	Varieties of lepidolite	P1	2	17/6		16/6	1/2	41/6	7/6	P1=A1+2AP/3 = Tri+5TP/12+MT/4
19		T1	2	21/8		23/8	1/2	29/4	3/4	T1=E1+23ET/24 = P1+5TP/24

* In order to differentiate from varieties of siderophyllite from No. 11 to 17, the abbreviation of the end-mineral siderophyllite (No. 4) is written as Fe-Eas or E, i.e. the abbreviation of Fe-analogue of eastonite.

† Foster (1960*a*) considered the micas having Li-occupancy of ~0.95−1.05 and octahedral occupancy of ~2.50−2.60 in half-cell formula as mixed forms.

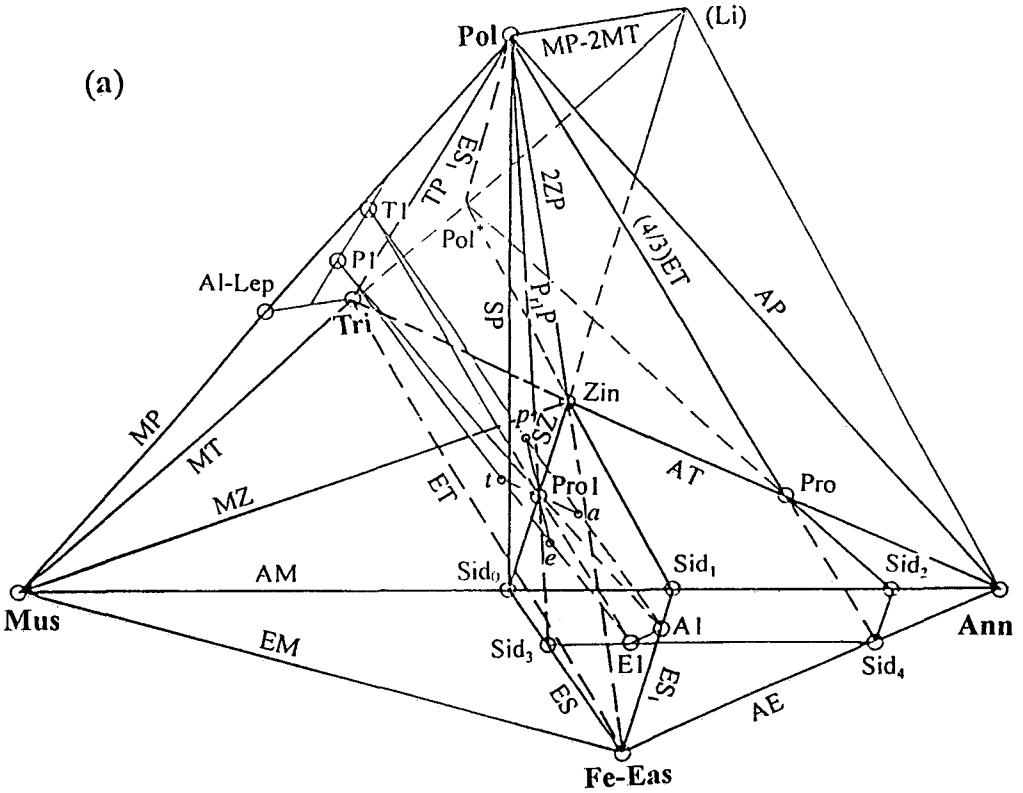

FIG. 1. A visual geometric frame for ideal Fe-Al-Li micas. (*a*) The geometric frame is composed of points, segments, vectors and planes, which express ideal mica points, mica joins, substitution vectors and substitution planes, respectively. Points Ann, Pol, Tri, Fe-Eas, Mus, Zin, Pro, Pro1, Al-Lep, Sid_0, Sid_1, Sid_2, Sid_3, Sid_4, A1, P1, E1 and T1 represent ideal Fe-Al-Li micas (Table 1). Segments linking pairs of ideal mica points, SP, $P_{ri}P$, ZP, AP, ET, MP, TP, MP−2MT, ES_1, ES, AE, EM, AT, AM, MT, SZ and MZ, represent both mica joins and substitution vectors without arrow (Table 2). Quadrilaterals APTE (Ann-Pol-Tri-Fe-Eas) and $APTE_{0.5}$ (A1-T1-P1-E1) and triangles AME (Ann-Mus-Fe-Eas), PMT (Pol-Mus-Tri), ATM (Ann-Tri-Mus) and PSE (Pol-Sid_0-Fe-Eas) are substitution planes. $APTE_{0.5}$ is parallel to APTE and passes through Pro1. Its two diagonals A1-P1 and E1-T1 and another two segments *ta* and *ep* intersect at point Pro1.

(also Sid_1 and Sid_2) (Table 1) are formed by muscovite replacement, $3Fe^{2+} \rightarrow 2Al_{VI} + \square_{VI}$. In order to differentiate Foster's Sid_0-Pol series, the siderophyllite (formula 6) as the Fe-analogue of eastonite is marked on Fe-Eas or E (Table 1), and Rieder's series, shown in formulae (4) or (5), on the Fe-Eas-Pol series.

Lapides *et al.* (1977) proposed two exchange equations for Fe-Li micas. One is equation (8) (The coefficient of $\square_{VI}$ of equation 1-8 in the original book was 1 instead of 0.5, which must be a typographic error) similar to equation (2) and the other (formula 9) is for Fe-Li micas with Li-occupancy <1.0−1.2 in a unit cell:

$$1.5R^{2+} + Al_{IV} + 0.5\square_{VI} + 2OH = 2Li + Si + 2F \quad (8)$$

$$R^{2+} + 0.8R^{3+}_{IV} + 0.2\square_{VI} = 1.2Li + 0.8Si \quad (9)$$

Equations (10) (Sun, 1988) and (11) (No. 5 in Table 3) are suggested to be essential replacements in the biotite-lepidolite series. Here, biotite as a low-Li member includes both siderophyllite and ferrobiotite (Sun and Yu, 1989*a*).

$$R^{3+}_{IV} + 3R^{2+}_{VI} \rightarrow Si + R^{3+}_{VI} + 2Li \quad (10)$$

$$2Al_{IV} + 4Fe^{2+} \rightarrow 2Si + Al_{VI} + 3Li \quad (11)$$

Assuming $\square_{VI}$ to be constant, Henderson *et al.* (1989) indicated that Al_{VI} occurs in the trend of

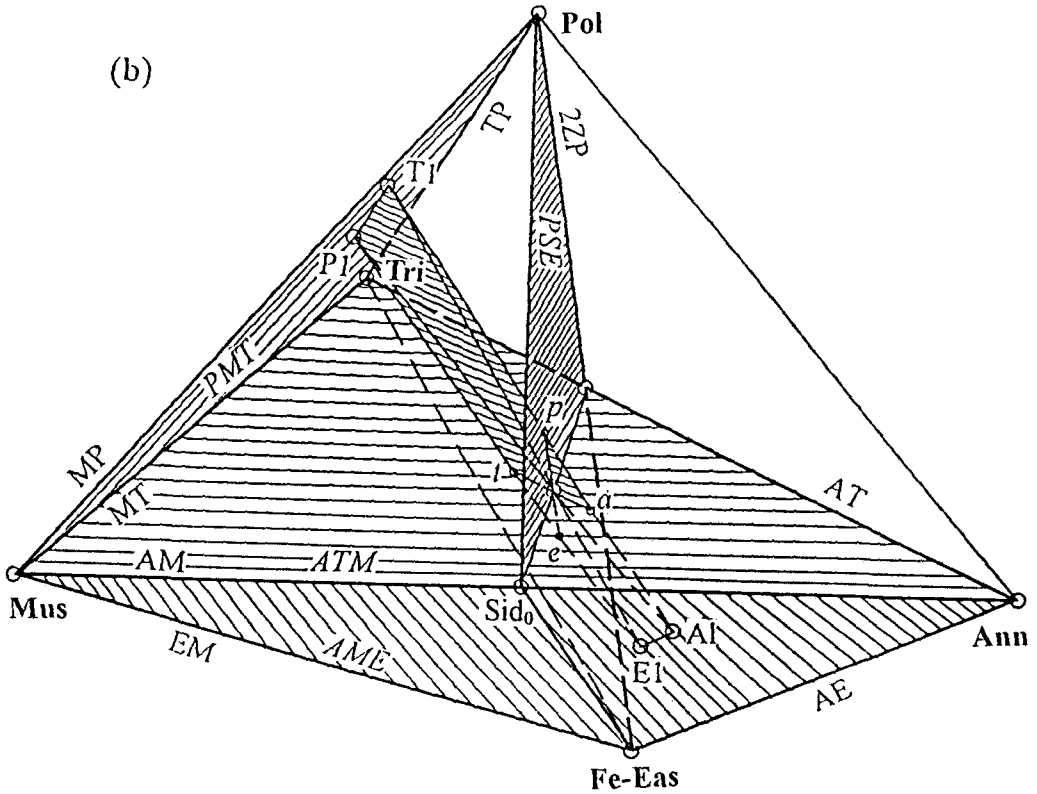

FIG. 1. (*contd.*) (*b*) The simplified frame for showing substitution planes PMT, ATM, AME, PSE and A1-T1-P1-E1 ($APTE_{0.5}$). A1E1, *ta*, *ep*, and P1T1 are intersection lines of A1-T1-P1-E1 with AME, ATM, PSE and PMT, respectively. They are parallel to AE, AT, ZP and TP, respectively.

unaltered Fe-Li micas as shown in (12), based on ion/electron-microprobe analyses of Li-micas in granites from SW England and France. Stone *et al.* (1997) argued that the high positive covariation reflected in formula (12) between Li and Al_{VI} is a result of a natural biotite trend and that formula (7) represents a Li-mica trend. Based on the nomenclature of Sun and Yu (1989*b*, Fig. 1), Stone's biotite trend is composed partly of protolithionite and partly of Mg-Fe micas ($Li<10\%_{\Sigma oct}$) with wide range of ratios for $(Fe^{2+}+Mn):Mg$ ($FM>2.70$) (where $FM = 6(Fe^{2+}+Mn)/(Fe^{2+}+Mn+Mg)$ for Mg-Fe micas (Sun and Yu, 1989*a*)).

$$2.2Li + 0.79Al_{VI} + 1.45Si = 3R^{2+}_{VI} + 1.45Al_{IV} \quad (12)$$

Equations and generalized formulae are effective for expressing replacements, but difficult to visualize and, it is especially difficult to show quantitatively, relations between replacements. Therefore, a frame expressing the substitution systems is proposed.

Substitution system and its geometric frame

The substitution system is the sum of mica compositions and their replacements. The geometric frame expressing the substitution system is composed of geometric elements: mica points representing ideal Fe-Al-Li micas, mica joins (segments), substitution vectors, and substitution planes. Figure 1*a* shows a 3-dimensional frame. This visual frame has its unique correspondent in 7-dimensional chemical composition space with the coordinate system (0; Si, Al_{IV}, Al_{VI}, Fe^{2+}, Li, $\square_{VI}$, K). The frame in hyperspace is abstract, but all geometric elements relative to ideal micas

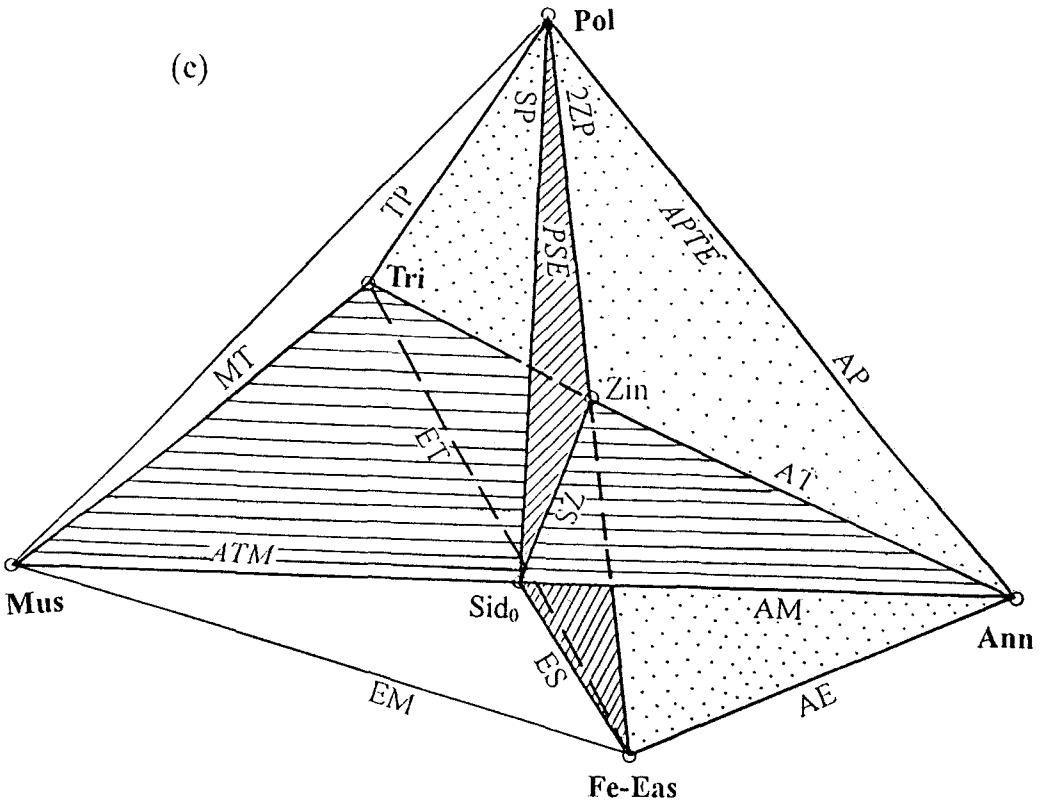

FIG. 1. (*contd.*) (*c*) The simplified frame for showing ATM, PSE and APTE. APTE intersects ATM along Ann-Tri (AT) and crosses PSE along Fe-Eas-Pol (2ZP). SZ is the intersection of PSE and ATM.

correspond to each other between abstract and visual frames.

Some quantities in the abstract frame, e.g. the angles between two planes or between plane and vector, are distorted in the polyhedron with a 3-dimensional orthogonal coordinate system (e.g. Černý and Burt, 1984, Fig. 9). In addition, these angles in the polyhedron will vary if its basis of coordinate system changes. Therefore, no coordinate system is attached in Fig. 1.

Ideal mica points

Some ideal Fe-Al-Li micas with unit-cell formula (13) are defined in Table 1. The apices of the visual frame (Fig. 1*a*,*b*,*c*) represent end-members of Fe-Al-Li micas: polylithionite (Pol), trilithionite (Tri), muscovite (Mus), annite (Ann) and siderophyllite (Fe-Eas). All points inside the frame represent intermediate ideal micas, e.g. zinnwaldite (Zin), varieties of siderophyllite (e.g. Sid_0 and Sid_4), protolithionite (Pro) and its variety, Pro1, etc.

$$K_2(Al_{VI},Fe^{2+},Li,\square_{VI})_6(Si,Al_{IV})_8O_{20}(OH,F)_4 \quad (13)$$

In the frame built in space (0; Si, Al_{IV}, Al_{VI}, Fe^{2+}, Li, $\square_{VI}$, K), suppose a mica point ε with coordinates (14) corresponds to an ideal mica with formula (13). Coordinates of ε must at least satisfy equations (15) to (17). So, the degree of freedom for the mica points is three, and ideal Fe-Al-Li micas can be expressed completely as a visual image as in Fig. 1. The 7-dimension coordinate system is also useful for studying replacement relationships among cations including interlayer K for natural micas, though the frame involves only tetrahedral and octahedral cations for ideal micas.

$$\varepsilon = (Si,Al_{IV},Al_{VI},Fe^{2+},Li,\square_{VI},K) \quad (14)$$
$$Al_{VI} + Fe^{2+} + Li + \square_{VI} = 6 \quad (15)$$
$$Si + Al_{IV} = 8 \quad (16)$$
$$4Si + 3(Al_{IV} + Al_{VI}) + 2Fe^{2+} + Li + K = 44 \quad (17)$$
$$K = 2 \quad (18)$$

Depending on the method of calculating the structural formulae, natural micas generally cannot simultaneously satisfy equations (15) to (18). So, natural micas can be expressed in hyperspace but do not fit exactly in any visual frame similar to Fig. 1.

Substitution vector and mica join

Substitution vector

A substitution vector is defined as the composition difference between two ideal micas. The substitution vector is a quantity having both magnitude (length) and direction. All substitution vectors are free vectors.

In the visual frame (Fig. 1*a*), substitution vector ET, for instance, is defined as the formula difference between micas in formula (19): Tri minus Fe-Eas. The ET direction indicates replacement relation among ions from Fe-Eas towards Tri. Its length is equal to that of segment Fe-Eas-Tri and is a measure showing the extent of replacement in the ET direction. The vector of segment Sid_4-Pol (Fig. 1*a*) is parallel to Fe-Eas-Tri and equal to (4/3)ET (No. 5 in Table 2) in length. Thus, Sid_4-Pol causes greater changes in the ET direction than Fe-Eas-Tri. Any segment inside the visual frame (Fig. 1*a*) is simply a substitution vector (arrowhead omitted). Some substitution vectors defined in such a way are listed in Table 2.

$$ET = Tri - Fe\text{-}Eas = (Al_{IV})_{-2}Fe^{2+}_{-4}Si_2Al_{VI}Li_3 \quad (19)$$

In the abstract frame, ET (No. 5 in Table 3) is defined as the coordinate difference (formula 20) between mica points: Tri minus Fe-Eas. The numbers in brackets in formula (20) are 'substitution components' of ET. The length of ET in hyperspace can be calculated with formula (A1.2) in the Appendix. Vectors defined here are listed in Table 3 and are analogous to those in Table 2.

Si	Al_{IV}	Al_{VI}	Fe^{2+}	Li	$\square_{VI}$	K	
(6	2	3	0	3	0	2) = Tri	
−(4	4	2	4	0	0	2) = Fe-Eas	
(2	−2	1	−4	3	0	0) = ET	(20)

Substitution vectors are equivalent to replacement equations (Table 3). For instance, ET can also be obtained by transposition of equation (21). Moving terms, together with their signs in the left side of equation (21), to the right side, the coefficients of the deformed equation constitute substitution vector (22), which is ET in 5-dimen-

TABLE 2. Definitions of substitution vectors without coordinates

No	Vector	Definition	Expression without coordinates
1	SP	SP = Pol−Sid	$(Al_{IV})_{-2}Fe^{2+}_{-3}(\square_{VI})_{-1}Si_2Li_4$
2	$P_{r1}P$	$P_{r1}P$ = Pol−Pro1 =3(Pol−Sid_3)/4	$(Al_{IV})_{-2}Fe^{2+}_{-5/2}(\square_{VI})_{-1/2}Si_2Li_3$
3	ZP	ZP = Pol−Zin =0.5(Pol−Fe-Eas)	$2[(Al_{IV})_{-1}Fe^{2+}_{-1}SiLi]$
4	AP	AP = Pol−Ann	$2[(Al_{IV})_{-1}Fe^{2+}_{-3}SiAl_{VI}Li_2]$
5	ET	ET = Tri−Fe-Eas =Pol−Pro=3(Pol−Sid_4)/4	$(Al_{IV})_{-2}Fe^{2+}_{-4}Si_2Al_{VI}Li_3$
6	MP	MP = Pol−Mus	$2[(Al_{IV})_{-1}(Al_{VI})_{-1}(\square_{VI})_{-1}SiLi_2$
7	TP	TP = Pol−Tri	$(Al_{IV})_{-2}(Al_{VI})_{-1}Si_2Li$
8	MP−2MT	MP −2MT	$2[(Al_{IV})_{-1}Li_{-1}Si\square_{VI}]$
9	ES_1	ES_1 = Sid_1−Fe-Eas	$2[(Al_{IV})_{-1}(Al_{VI})_{-1/3}(\square_{VI})_{1/3}Si]$
10	ES	ES = Sid_0−Fe-Eas	$(Al_{IV})_{-2}Fe^{2+}_{-1}Si_2\square_{VI}$
11	AE	AE = Fe-Eas−Ann = Mus−Phe	$2[Si_{-1}Fe^{2+}_{-1}Al_{IV}Al_{VI}]$
12	EM	EM = Mus−Fe-Eas	$2[(Al_{IV})_{-1}Fe^{2+}_{-2}SiAl_{VI}\square_{VI}]$
13	AT	AT =Tri−Ann	$3[Fe^{2+}_{-2}Al_{VI}Li]$
14	AM	AM = Mus−Ann	$2[Fe^{2+}_{-3}(Al_{VI})_2\square_{VI}]$
15	MT	MT = Tri−Mus	$(Al_{VI})_{-1}(\square_{VI})_{-2}Li_3$
16	SZ	SZ = Zin−Sid	$Fe^{2+}_{-1}(\square_{VI})_{-1}Li_2$
17	MZ	MZ = Zin−Mus	$2[(Al_{VI})_{-1}(\square_{VI})_{-1}Fe^{2+}Li]$

TABLE 3. Substitution vectors with coordinates and substitution equations

Vector No.		Substitution components Si	Al_{IV}	Al_{VI}	Fe^{2+}	Li	$\square_{VI}$	K	Substitution equation or generalized formula
1	SP	2	−2		−3	4	−1		$3nFe^{2+}+2nAl_{IV} \rightarrow 4nLi+2nSi$ (1) or $1.5R^{2+}+Al_{IV}+0.5\square_{VI} \rightarrow 2Li + Si$ (2)
2	$P_{r1}P$	2	−2		−5/2	3	−1/2		$R^{2+}_{VI}+0.8R^{3+}{}_{IV}+0.2\square_{VI} \rightarrow 1.2Li+0.8Si$ (2)
3	ZP	2	−2		−2	2			$K_2Fe^{2+}_XLi_{4-X}(Al,Fe^{3+})_2Al_XSi_{8-x}O_{20}(OH,F)_4$ (3) or $K_2R^{2+}_XLi_{4-x}R^{3+}_2R^{3+}_xSi_{8-X}O_{20}(OH,F)_4$ (4), $0 \leqslant X \leqslant 4.0$
4	AP	2	−2	2	−6	4			$R^{3+}_{IV}+3R^{2+}_{VI} \rightarrow Si+R^{3+}_{VI}+2Li$ (5)
5	ET	2	−2	1	−4	3			$2Al_{IV}+4Fe^{2+} \rightarrow 2Si+Al_{VI}+3Li$ (7)
6	MP	2	−2	−2		4	−2		nLi+0.5nSi +0.5nOct.Site → nAl (1)
7	TP	2	−2	−1		1			$2Al_{IV}+Al_{VI} \rightarrow 2Si+Li$
8	MP−2MT	2	−2			−2	2		$Al_{IV}+Li \rightarrow Si+\square_{VI}$ (3)
9	ES_1	2	−2	−2/3			2/3		$3Al_{IV}+Al_{VI} \rightarrow 3Si+\square_{VI}$ (3)
10	ES	2	−2		−1		1		$2Al_{IV}+R^{2+}_{VI} \rightarrow 2Si+\square_{VI}$ (3)
11	AE	−2	2	2	−2				$Si+Fe^{2+} \rightarrow Al_{IV}+Al_{VI}$ (Fe^{2+}-Tschermak)
12	EM	2	−2	2	−4		2		$Al_{IV}+2Fe^{2+} \rightarrow Si+Al_{VI}+\square_{VI}$
13	AT			3	−6	3			$2Mg \rightarrow Al_{VI}+Li$ (6)
14	AM			4	−6		2		$3R^{2+}_{VI} \rightarrow 2R^{3+}_{VI}+\square_{VI}$
15	MT			−1		3	−2		$0.5n$Al → $1.5n$Li + nOct. Site (1)
16	SZ				−1	2	−1		$R^{2+}_{VI}+\square_{VI} \rightarrow 2R^{+}_{VI}$ (3)
17	MZ			−2	2	2	−2		$Al_{VI}+\square_{VI} \rightarrow Fe^{2+}+Li$ (2)

Notes: (1) Foster, (1960*a*); (2) Lapides *et al.* (1977); (3) Rieder (1970); (4) Stone *et al.* (1988); (5) Sun (1988); Ginzburg and Berkhin (1953); (7) This paper.

sional composition subspace (0; Si, Al_{IV}, Al_{VI}, Fe^{2+}, Li).

$$2Al_{IV} + 4Fe^{2+} \rightarrow 2Si + Al_{VI} + 3Li \quad (21)$$

Si Al_{IV} Al_{VI} Fe^{2+} Li

(2 −2 1 −4 3) (22)

Suppose formula (23) is the generalized substitution vector V. Here, vcn is the $\square_{VI}$ component of V, C^{ei}_{Ri} the cation R^{ei}_i component, and e_i the valence of R^{ei}_i (i =1, 2, ...*n*). Component Sum (CS) and Valence Sum (VS) of V are defined as formulae (24) and (25). When R^{ei}_i is only octahedral cations, CS and VS are marked as CS_{VI} and VS_{VI}. The CS and VS are substitution parameters, which together with the components of vector V express the substitutional characters of V. Not all vectors in hyperspace are substitution vectors in the same way as not all points are ideal or actual micas. CS + vcn = 0 and VS = 0 are the restriction of substitution vectors.

$$V = (C^{e1}_{R1}, C^{e2}_{R2}, \ldots C^{en}_{Rn}, \text{vcn}) \quad (23)$$

$$CS = \Sigma\, C^{ei}_{Ri} \quad (24)$$

$$VS = \Sigma\, e_i \cdot C^{ei}_{Ri} \quad (25)$$

Mica join and its composition expression

The term segment or mica join is used to express a mica combination with composition restrictions along it. According to the definition of substitution vector, the composition expression of the Pro-Pol join for both frame (Fig. 1*a*) and hyperspace is Pro + *t*(Pol −Pro); here Pol −Pro = ET (Table 2). The extension line of this join intersects plane AME (see next subsection and Fig. 1*a*) at point Sid_4 and Sid_4=Pro + t_{AME}ET. The t_{AME}-value can be obtained by two methods. The first is the plane-equation method suitable for any kind of plane, i.e. computing with formula (A4.4) in the Appendix. The second is the plane-constant method only for fundamental types of planes. In the case of AME, the constant composition is Li = 0 (Table 4) and t_{AME}= −1/3. The Sid_4 obtained is exactly the same as No. 15 in Table 1, and the Sid_4-Pol join (Fig. 1*a*) has its composition expression Pro +*t*ET ($-1/3 \leqslant t \leqslant 1$).

Both the mica join and its expression are used for describing a mica series. For example, Rieder (1968, 1970) described his Fe-Li mica series as

the join of Fe-Eas-Pol instead of the join of Fe-Eas-Tri or Ann-Tri. The Fe-Eas-Pol join has its expression Fe-Eas $+2x$ZP $(0 \leqslant x \leqslant 1)$, which is equivalent to formula (4).

Fundamental type of substitutions and representative plane

A substitution plane is determined by linear combination of a couple of linearly independent substitution vectors. The substitution plane is a set of substitution vectors and ideal mica points with certain characters. The common substitutional character of all vectors on a substitution plane and the common compositional character of all mica points on it are called the substitutional and compositional characters of the plane respectively (Table 4). Some plane equations are given in the Appendix (equation (A1.4) and Table A1).

All substitution planes parallel to each other are taken as a type of plane expressing a type of substitution. A type of plane with a constant value of substitution parameter is called the 'fundamental type'. The different planes of each fundamental type are of the same constant parameter but different constant composition (i.e. different compositional character). In the visual or abstract frame, there are only five fundamental types, which are represented by planes ATM, PSE, APTE (or $APTE_{0.5}$), PMT and AME, shown in Fig. 1*b,c*. Triangle APM connecting Ann, Pol and Mus (not shown in Fig. 1), for instance, is not a fundamental type of plane for it has no constant parameter. There are innumerable non-fundamental-type planes in the frame. The generalized formulae and constant quantities describing the characters of each fundamental plane are listed in Table 4.

Al_{IV} = 2 ATM and VS_{VI} = 0 substitutions

ATM (Fig. 1*b,c*) is defined as triangle Ann-Tri-Mus. The type of substitution represented by ATM is that all substitutions are confined within the octahedral sheet, i.e. $VS_{VI} = \Sigma e_i.C^{ei}_{RVI} = 0$. Here, C^{ei}_{RVI} is the octahedral-cation-R^{ei}_{VI} component of substitution vectors. Al_{IV}=2 is the compositional character of ATM, i.e. all mica points on ATM have the same composition $Si_sAl_a = Si_6Al_2$. Points of micas with s:a>6:2 (e.g. polylithionite) sit above ATM in the visual frame, and those with s:a<6:2 (e.g. Fe-eastonite) below it. Many natural micas, such as protolithionite, siderophyllite, biotite, phlogopite and muscovite, plot near ATM in hyperspace, due to $|X|<0.5$, generally, for their $Si_{6+X}Al_{2-X}$.

$Pol^* = Mus + (4/3)MT$ is beyond the composition of an ideal Fe-Al-Li mica. If Pol^* is considered just as projection of Pol on the extension line of the Mus-Tri side, triangle Li-Mus-Ann (Fig. 1*a*) becomes Foster's (1960*a*) Li-mica triangle.

Al_{VI} =2 PSE and Al_{VI}-constant substitutions

PSE (Fig. 1*b,c*) is triangle Pol-Sid_0-Fe-Eas. Its substitutional character is that substitutions do not change Al_{VI}-occupancy ($C_{Al_{VI}} = 0$). The length of ZP is half as long as its Fe-Eas-Pol side. PSE has constant composition, $Al_{VI} = 2$. Mica points with $Al_{VI}<2$ plot on the right side of PSE in the visual frame, and those with $Al_{VI}>2$ on the left side.

TABLE 4. Substitutional and compositional characters of substitution planes: their generalized formulae and associated constant quantities

Substitution plane	Substitutional character		Compositional character	
	General replacement	Constant parameter	General composition	Constant composition
ATM	aAT +bSZ	VS_{VI} =0	Ann +aAT +bSZ	Al_{IV} =2
PSE	aZP +bSZ	C_{AlVI} =0	Fe-Eas +aZP +bSZ	Al_{VI} =2
APTE	aAT +bAE	CS_{VI} =0	Ann +aAT +bAE	$\square_{VI}$ =0
PMT	aMT +bMP	$C_{Fe^{2+}}$=0	Mus +aMT +bMP	Fe^{2+} =0
AME	aAM +bAE	C_{Li} =0	Ann +aAM +bAE	Li =0
$APTE_{0.5}$	aAT +bAE	CS_{VI} =0	Pro1+aAT +bAE	$\square_{VI}$ =0.5

Note: The combinations of three independent vectors, e.g. AT, AE and SZ, are enough to express all the generalized formulae: ZP = (2/3)AT −AE, MT = −(1/3)AT +2SZ, and MP = −AE +2SZ.

The generalized mica formula of PSE is Fe-Eas +aZP+bSZ, which is equivalent to formula (26). When Y=4−X, formula (26) becomes a generalized formula, the same as formula (4), an explanation of the Fe-Li mica series.

$$K_2Al_2Fe^{2+}{}_XLi_YSi_{12-2X-Y}Al_{2X+Y-4}O_{20}(OH,F)_4 \quad (26)$$

$\square_{VI}$ = 0 APTE and CS_{VI} =0 substitutions

APTE (Fig. 1*c*) is the planar quadrilateral Ann-Pol-Tri-Fe-Eas. The substitutional character of planes represented by APTE is $CS_{VI} = \Sigma C^{ei}_{RVI} = 0$, i.e. substitutions do not change the numbers of octahedral vacancy. All ideal micas with compositional character $\square_{VI} = 0$ are coplanar on APTE. All $CS_{VI} = 0$ planes in Fig. 1 are $\square_{VI} \geqslant 0$, planes parallel to APTE and occurring on its left side.

Quadrilateral A1-T1-P1-E1 passing through Pro1 (Fig. 1*a*,*b*) is a part of $APTE_{0.5}$ (Table 4). Its diagonals A1-P1 and E1-T1 fit well to the trend of natural Fe-Li micas (Sun and Yu, in prep.).

Fe^{2+}-free PMT and Fe^{2+}-constant substitutions

PMT (Fig. 1*b*) is the triangle Pol-Mus-Tri and is equivalent to Muñoz's (1968) triangle muscovite-trilithionite-polylithionite. Any replacement vector without Fe^{2+} ($C_{Fe^{2+}} = 0$) is parallel to PMT. Ideal Fe-Al-Li micas with $Fe^{2+} = 0$ and points on PMT correspond to each other. Vectors MT and MP are the most important replacements for Al-Li micas (Foster, 1960*a*; Chaudhry and Howie, 1973).

Li-free AME and Li-constant substitutions

AME (Fig. 1*b*) is the triangle Ann-Mus-Fe-Eas. AME can be enlarged as the planar quadrilateral Ann-Al-Cel-Mus-Fe-Eas because aluminoceladonite (Al-Cel, No. 6 in Table 1, not shown on Fig. 1) is also Li-free. The constant parameter of substitutions represented by AME is $C_{Li} = 0$.

AME represents Fe-Al micas that occur in Li-deficient and f_{O_2}-low environments of crystallization. Simultaneous replacements of AM and AE result in a break of continuity of variation in Al-distribution between tetrahedral and octahedral sheets (Sun and Yu, 1989*b*, Fig. 2 and 1984, Fig. 4).

Discussion

The frame can express the entire substitution system

Not all substitution vectors and substitution planes of ideal micas in the frame are shown. In fact, the planes mentioned above, e.g. AME, can be enlarged, and not all bounding planes (e.g. $Al_{IV} = 0$ and $Al_{IV} = 4$ planes) are shown in Fig. 1. However, for our purpose, both visual and abstract frames have expressed the entire substitution system of Fe-Al-Li micas as formula (13). The reasons are as follows. (1) All of the five fundamental types of substitutions have been expressed in the frame. (2) Any ideal mica and any segment expressing the Fe-Li mica series [e.g. Fe-Eas + $2x$ZP $(0 \leqslant x \leqslant 1)$] have been restricted by Li-free AME and Fe-free PMT. (3) With the combination of any three vectors on ATM, APTE and PSE, respectively, e.g. formula (27), all ideal micas and their replacement relations can be expressed simply.

$$t(AT - xAE + kSZ) \quad (27)$$

The angles (Table 5) between each pair of planes are calculated based on normal vectors of planes (Table A1 in the Appendix) with equation (A3.4). The angles of ATM relative to PSE and APTE are as large as $\pm 69.7°$, and the angle between PSE and APTE is 62.2°. The large angles imply great differences in substitutional characters between these definitions.

TABLE 5. Angles between fundamental substitution planes in composition space with the coordinate system (0; Si, Al_{IV}, Al_{VI}, Fe^{2+}, $\square_{VI}$, Li, K)

Substitution plane	APTE $CS_{VI} = 0$	PSE $Al_{VI} = 2$	ATM $VS_{VI} = 0$	PMT $Fe^{2+} = 0$	AME $Li = 0$
APTE	0°				
PSE	62.2°	0°			
ATM	69.7°	−69.7°	0°		
PMT	76.6°	38.0°	85.4°	0°	
AME	38.0°	76.6°	−85.4°	−73.1°	0°

Four types of series proposed

Based on the geometric position of the segment (or vector) relative to ATM, APTE and PSE, six expressions proposed for Fe-Li mica series are divided into four types.

Type I: the intersection of ATM and APTE (Fig. 1c)

In this case, the essential replacement is AT (cf. Ginzburg and Berkhin, 1953), which has substitution characters of ATM and APTE, i.e. $CS_{VI} = 0$ and $VS_{VI} = 0$.

Type 2: the joins of Sid_0-Pol and Prol-Pol on PSE (Fig. 1a)

The direction vectors of these two joins (or their essential replacements) are SP and $P_{r1}P$ (cf. Foster, 1960*a*, Lapides *et al.*, 1977). In this case, $Al_{VI} = 2$ in micas.

Type 3: intersection of PSE and APTE (Fig. 1c)

In this case, the essential replacement is ZP (cf. Rieder, 1970; Stone *et al.*, 1988), which keeps constant compositions of PSE and APTE, $Al_{VI} = 2$ and $\square_{VI} = 0$, concurrently.

Type 4: substitution vectors AP and ET on APTE (Fig. 1a)

These are defined as the essential replacements (Sun, 1988 and No. 5 in Table 3). Both have constant parameter $CS_{VI} = 0$, i.e. $\square_{VI}$ is a constant component during replacement.

The replacements from AT (type 1) to SP, $P_{r1}P$, and ZP have been changed enormously: the angles (Table 6) of AT relative to the latter are the greatest, >45°. SP and $P_{r1}P$ as the same type (type 2) have the smallest angles, 7.9°. ZP (type 3) shows only slightly larger angles to SP and $P_{r1}P$, 19.4° and 11.5° respectively. AP is a special replacement, which, compared with replacements of the other three types, has intermediate angles (from 23 to 29°). ET and AP as the same type (type 4) are similar to each other (9.6°), though ET is slightly closer to ZP, SP and $P_{r1}P$ but further from AT than AP.

TABLE 6. Angles between supposed essential replacements in composition space with coordinate system (0; Si, Al_{IV}, Al_{VI}, Fe^{2+}, $\square_{VI}$, Li, K)

Vectors	ET	AP	ZP	$P_{r1}P$	SP	AT
ET	0°					
AP	9.6°	0°				
ZP	19.4°	29.0°	0°			
$P_{r1}P$	17.2°	25.5°	11.5°	0°		
SP	19.7°	25.8°	19.4°	7.9°	0°	
AT	32.8°	23.3°	52.2°	47.6°	45.6°	0°

Fe-Li mica series have no more than five possible types

Formula (27) can not express the mica series without restriction, simply. All studies on Fe-Li mica series have an implicit assumption: to find one or two fixed quantities of Al_{IV} (Si), Al_{VI}, and $\square_{VI}$ in the trend of natural Fe-Li micas. Therefore, the types of segments that possibly express the Fe-Li mica series have only six: three types of segments are on ATM, APTE or PSE, and the others the intersection lines between each pair of planes. Besides the previous four types, the two other types are the intersection of ATM and PSE (the Sid_0-Zin join) and segments on ATM (Fig. 1*a*,*c*).

The Sid_0-Zin join, however, does not end at Tri. If PSE is translated to pass through Tri, the Li-free end of the new intersection becomes a dioctahedral mica, $K_2Al_3Fe^{2+}{}_{3/2}(\square_{VI})_{3/2}Si_6Al_2O_{20}(OH)_4$. Therefore, the intersection between ATM and PSE can not define the Fe-Li mica series.

All vectors on ATM have a formula $V_{ATM} = AT - yAM$. When $y = 0$, $V_{ATM} = AT$, which is the type 1 mentioned above. When $y = 1/2$, V_{ATM} becomes the direction vector of segment Sid_0-Tri. When $y > 1/2$, the points on Li-free side of ATM are dioctahedral because their octahedral occupancies are <5.0 (according to the definition of Rieder *et al.*, 1999). So, V_{ATM} ($0 < y \leqslant 1/2$) may also be a definition of the Fe-Li mica series and is called ATM-type or Al_{IV}-constant type.

Consequently, the number of possible types of series definitions is five at most. All possible types of the series have been proposed except the ATM-type.

Restriction of the expression $t(AT - xAE + kSZ)$

Six essential replacements, formulae (28) to (33), correspond to six supposed definitions of the Fe-Li mica series.

$$AT = AT + 0AE \quad (28)$$
$$AP = (4/3)\,(AT - 0.75AE) \quad (29)$$
$$ET = AT - AE \quad 30)$$
$$ZP = (2/3)\,(AT - 1.5AE) \quad (31)$$
$$SP = (2/3)\,((AT - 1.5AE) + 1.5SZ) \quad (32)$$
$$P_{r1}P = (4/3)\,((AT - 1.5AE) + 0.75SZ) \quad (33)$$

The previously mentioned four types of the series imply four different kinds of restrictions of x- and k-values in the generalized expression (27). AT is the common component of the six essential replacements. The dispute on restrictions is whether or not octahedral AT-replacement needs to be accompanied simultaneously by replacement of Si for Al_{IV} ($x = 0$), by filling octahedral vacancy ($k = 0$), and by increase in Al_{VI} to balance the AE-adjustment ($x = 1.5$).

Reliability of the definitions of the Fe-Li mica series

What compositional characters do natural micas have? What kind of vector best fits the composition trend of natural micas? The definitions of the Fe-Li mica series need to be compared with the spatial positions of geometric elements of the frame with natural Fe-Li micas and with the trend variation of micas. The frame is a datum system for the comparison, and the formulae listed in the Appendix are a means for this comparison. However, further work is needed in order to determine quantitatively the trend variation of natural Fe-Li micas (Sun and Yu, in prep.) before we can make that comparison.

Conclusions

A new geometric frame with both visual and abstract forms has been set up for expressing the entire substitution scheme of the Fe-Al-Li micas. The frame is used for two purposes: to differentiate proposed definitions of the Fe-Li mica series and to determine quantitatively the range of the Fe-Li mica series.

For the former, it has been proved that the main issue for the Fe-Li mica series is whether the replacement of Li for Fe^{2+} needs the balance of Al_{IV}, Al_{VI} or $\square_{VI}$. For the latter, a set of formulae determining spatial positions and relations of geometric elements in multi-composition space has been suggested. The frame, together with the calculation formulae, is a datum system to measure the compositional and substitutional characters of natural Fe-Li micas quantitatively.

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Appendix

The following discussion is carried out in a 7-dimensional chemical composition space with the coordinate system (0; Si, Al_{IV}, Al_{VI}, Fe^{2+}, Li, $\square_{VI}$, K).

Equation of substitution plane

Suppose P_N is a substitution plane in composition space. Plane P_N has a normal vector N (A1.1) with length L_N (A1.2). A mica point ε on P_N represents a mica with composition (A1.3), which must satisfy equation (A1.4) of plane P_N.

$$N = (n_1, n_2, n_3, n_4, n_5, n_6, n_7) \quad \text{(A1.1)}$$

$$L_N = \left(\sum_{i=1}^{7} n_i^2\right)^{1/2} \quad \text{(A1.2)}$$

$$\varepsilon = (Si, Al_{IV}, Al_{VI}, Fe^{2+}, Li, \square_{VI}, K) \quad \text{(A1.3)}$$

$$n_1Si + n_2Al_{IV} + n_3Al_{VI} + n_4Fe^{2+} + n_5Li + n_6\square_{VI} + n_7K + C = 0 \quad \text{(A1.4)}$$

The K-value of any mica (ε) in plane P_N is = 2. C in (A1.4) is called the constant of P_N. The direction numbers of N constitute the coefficients of variables in equation (A1.4). Equation coefficients and constant C together with length L_N of N are listed in Table A1 for some planes explained in the text. Plane APM (Ann-Pol-Mus) is not shown in Fig. 1*b*,*c*.

Distances of mica points to a substitution plane

Suppose a group of *m* mica analyses is expressed as *m* points in composition space. The distance of point S_j (A2.1) to plane P_N (A1.4) is d_j (A2.2). The mean and variance of distances of the *m* points to P_N are $\bar{d}$ (A2.3) and υ (A2.4).

$$S_j = (s_{j1}, s_{j2}, s_{j3}, s_{j4}, s_{j5}, s_{j6}, s_{j7}) \quad \text{(A2.1)}$$

$$d_j = \left|\sum_{i=1}^{7} n_i s_{ji} + C\right| / L_N \quad \text{(A2.2)}$$

TABLE A1. The coefficients n_i and constant C of plane equation (A1.4), and length L_N of normal vector N for each substitution plane

Substitution Plane	Normal vector N							Constant C	Length L_N
	n_1	n_2	n_3	n_4	n_5	n_6	n_7		
ATM	10.0	−10.0	−3.0	−1.0	1.0	3.0	0.0	−34.0	$2(55)^{1/2}$
PSE	− 6.0	6.0	15.0	−17.0	−5.0	7.0	0.0	38.0	$2(165)^{1/2}$
APTE	6.0	−6.0	7.0	−5.0	−17.0	15.0	0.0	6.0	$2(165)^{1/2}$
$APTE_{0.5}$	6.0	−6.0	7.0	−5.0	−17.0	15.0	0.0	−16.0	$2(165)^{1/2}$
PMT	2.0	−2.0	17.0	−31.0	9.0	5.0	0.0	−86.0	$2(341)^{1/2}$
AME	−2.0	2.0	5.0	9.0	−31.0	17.0	0.0	−46.0	$2(341)^{1/2}$
APM	38.0	−38.0	−7.0	5.0	−27.0	29.0	0.0	182.0	$2(1133)^{1/2}$

$$\bar{d} = \sum_{i=1}^{7} d_j/m \quad \text{(A2.3)}$$

$$\upsilon = \sum_{j=1}^{m} (d_j - \bar{d})^2/(m-1) \quad \text{(A2.4)}$$

The angles between substitution vectors and planes

Suppose V (A3.1) and W (A3.2) are two vectors (substitution vector or eigenvector) with length L_V and L_W respectively, which are expressed in the same way as (A1.2). Arcsin β (A3.3) is the angle between vector V and plane P_N, and arccos α (A3.4) is that between V and W. If V and W are the normal vectors of the substitution planes P_V and P_W, arccos α (A3.4) shows the size of the angle between P_V and P_W. When $\alpha > 90°$, the angle between two planes is expressed as $\alpha - 180°$.

$$V = (v_1, v_2, v_3, v_4, v_5, v_6, v_7) \quad \text{(A3.1)}$$
$$W = (w_1, w_2, w_3, w_4, w_5, w_6, w_7) \quad \text{(A3.2)}$$

$$\text{Sin } \beta = \sum_{i=1}^{7} | n_i v_i | /(L_N L_V) \quad \text{(A3.3)}$$

$$\text{Cos } \alpha = \sum_{i=1}^{7} V_i w_i/(L_V L_W) \quad \text{(A3.4)}$$

Coordinates of the intersection point between the mica join and plane P_N

Suppose mica join (straight line) Y_V (A4.2) is parallel to vector V (A3.1), passes through mica point Q (A4.1), and intersects plane P_N (A1.4) at point x. Line Y_V has parameter t. Coordinates of intersection point x, Y_x, can be computed with equation (A4.3).

$$Q = (\mathbf{q_1}, \mathbf{q_2}, \mathbf{q_3}, \mathbf{q_4}, \mathbf{q_5}, \mathbf{q_6}, \mathbf{q_7}) \quad \text{(A4.1)}$$
$$Y_V = Q + t\,V \quad \text{(A4.2)}$$
$$Y_x = Q + t_P V \quad \text{(A4.3)}$$

Here,

$$t_P = -(\sum_{i=1}^{7} n_i q_i + C)/\sum_{i=1}^{7} n_i v_i \quad \text{(A4.4)}$$

Distance between a mica point and the mica join $Y_V = Q + tV$

Suppose mica points Q and R have compositions (A4.1) and (A5.1), vector V (A3.1) has length L_V. The vector from Q to R, QR, has length L_{QR} (A5.2) and the angle between QR and V is arccos α (A5.3). The distance of R to mica join $Y = Q + tV$ is L (A5.4 or A5.5).

$$R = (\mathbf{r_1}, \mathbf{r_2}, \mathbf{r_3}, \mathbf{r_4}, \mathbf{r_5}, \mathbf{r_6}, \mathbf{r_7}) \quad \text{(A5.1)}$$

$$L_{QR} = [\sum_{i=1}^{7} (\mathbf{q_i} - \mathbf{r_i})^2]^{1/2} \quad \text{(A5.2)}$$

$$\cos \alpha = [\sum_{i=1}^{7} \mathbf{v_i}(\mathbf{q_i} - \mathbf{r_i})]/(L_V L_{QR}) \quad \text{(A5.3)}$$

$$L = L_{QR} \sin \alpha \quad \text{(A5.4)}$$

$$L = \{L_{QR}^2 - [\sum_{i=1}^{7} \mathbf{v_i}(\mathbf{q_i} - \mathbf{r_i})]^2/L_V^2\}^{1/2} \quad \text{(A5.5)}$$