

Correlation of Diocahedral Potassium Micas on the Basis of Their Charge Relations

GEOLOGICAL SURVEY BULLETIN 1036-D



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By MARGARET D. FOSTER

A CONTRIBUTION TO GEOCHEMISTRY

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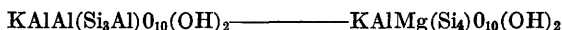
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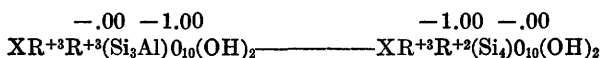
By MARGARET D. FOSTER

ABSTRACT

In formulas derived from analyses of many varieties of dioctahedral potassium micas the number of Si ions in the tetrahedral group lies between three and four, that is, the micas represented by the formulas are all members of a trisilicic-tetrasilicic series like that suggested by Schaller to interpret the composition of high silica sericites



Generalization of these formulas by substitution of the symbols R^{+3} and R^{+2} for Al and Mg permits extension of this series to interpret the composition of dioctahedral potassium micas containing other cations than Al and Mg, such as ferric and ferrous iron, vanadium, chromium and manganese,



In this series constancy of charge and potassium content require that increase in silicon ion content be accompanied by an equivalent increase in divalent ion content and by a decrease in aluminum ion content equivalent to the increase in silicon and divalent ion content. In the trisilicic end-member the charge on the structural layers responsible for holding potassium is due entirely to the proxying of trivalent aluminum for one-fourth of the tetravalent silicon in the tetrahedral group; in the tetrasilicic end-member it is due to the proxying of bivalent ions, ion for ion, for one-half the trivalent ions in the octahedral group. From the trisilicic to the tetrasilicic end-members there is, therefore, a shift in the seat of the charge from the tetrahedral to the octahedral group. In intermediate members of the series the charge is due partly to replacement of aluminum for silicon in the tetrahedral group and partly to replacement of bivalent for trivalent ions in the octahedral group. It is this tetrahedral-octahedral charge relationship that is herein suggested as a basis for the classification and correlation of the dioctahedral potassium micas.

INTRODUCTION

The dioctahedral potassium micas vary greatly in composition, particularly in their relative content of silicon, aluminum, ferric and ferrous iron, and magnesium. Some also contain chromium, manganese, and vanadium. The composition of some of the high-silica sericites and their relation to muscovite was explained by Schaller

(1950) but the composition and relationships of other dioctahedral potassium micas have not been fully interpreted.

The great difference in composition found among these micas is the result of isomorphous replacement. The position that a cation takes in the mica structure is determined by its size, not by its valence. For this reason the composition of the outer or tetrahedral layers is usually comparatively simple, as only silicon and aluminum normally are small enough to take tetrahedral coordination. On the other hand, a number of cations, such as Al, Fe^{+3} , Cr^{+3} , Mn^{+3} , V^{+3} , Mg, Fe^{+2} , Mn^{+2} , and Li, have the proper size to take octahedral coordination. As several of these may occur in the same specimen, the composition of the octahedral layer may be quite complex.

Although the size of a cation determines the position it can take in the structure, its valence in this position is of importance with respect to the charge in the structural layers. Aluminum proxying for silicon in tetrahedral layers gives rise to a charge equivalent to the amount of aluminum in the tetrahedral layers. Octahedral layers that contain some divalent ions in the two positions occupied in dioctahedral micas have a charge equivalent to the divalent ions present. The seeming complexity in the composition of dioctahedral potassium micas can, therefore, be simplified if the composition is considered in terms of the position and valence of the constituent ions. It is the purpose of this paper to show that these micas can be classified and correlated on the basis of the relation between the charges on their tetrahedral and octahedral layers.

PYROPHYLLITE-MUSCOVITE RELATIONSHIP

The general structural scheme of the micas and related minerals was formulated by Pauling (1930), who showed that these minerals consist of composite layers made up of a sequence of atomic sheets or layers along the pseudohexagonal axis. For pyrophyllite and muscovite the succession of atomic layers is as follows:

	Pyrophyllite	Muscovite	
Tetrahedral layer.	$\begin{cases} 6 \text{ O}^- \\ 4 \text{ Si}^{+4} \end{cases}$	$\begin{cases} 6 \text{ O}^- \\ 3 \text{ Si}^{+4} + \text{Al}^{+3} \end{cases}$	} Charge (-2)
Octahedral layer.	$\begin{cases} 4 \text{ O}^- + 2(\text{OH}, \text{F})^- \\ 4 \text{ Al}^{+3} \\ 4 \text{ O}^- + 2(\text{OH}, \text{F})^- \end{cases}$	$\begin{cases} 4 \text{ O}^- + 2(\text{OH}, \text{F})^- \\ 4 \text{ Al}^{+3} \\ 4 \text{ O}^- + 2(\text{OH}, \text{F})^- \end{cases}$	
Tetrahedral layer.	$\begin{cases} 4 \text{ Si}^{+4} \\ 6 \text{ O}^- \end{cases}$	$\begin{cases} 3 \text{ Si}^{+4} + \text{Al}^{+3} \\ 6 \text{ O}^- \end{cases}$	

		2 K^+	} Charge (+2)
	$\begin{cases} 6 \text{ O}^- \\ 4 \text{ Si}^{+4} \end{cases}$	$\begin{cases} 6 \text{ O}^- \\ 3 \text{ Si}^{+4} + \text{Al}^{+3} \end{cases}$	

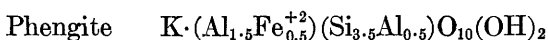
In pyrophyllite the succession of atomic layers forms a neutral structure; the number of negative charges equals the number of positive charges. But in muscovite the composite layer has a negative charge due to the replacement of one-fourth of the tetravalent silicon ions in the tetrahedral layers by trivalent aluminum. To achieve a neutral structure, positive ions (such as K) must be introduced. There is room for such ions in the pockets formed by the six oxygen ions on the top of the composite layer below and on the bottom of the composite layer above.

As the negative charge, which is neutralized by potassium, is due to the replacement of one-fourth of the silicon ions by aluminum in the tetrahedral layers, the seat of the charge in muscovite may be said to be in the tetrahedral layers.

The chemical composition of pyrophyllite is represented by the formula, $\text{Al}_2\text{Si}_4\text{O}_{10}(\text{OH})_2$ and that of muscovite by the formula, $\text{K} \cdot \text{Al}_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH})_2$, although these formulas do not represent the composition of the unit cells of these minerals.

TRISILICIC-TETRASILICIC SERIES

Winchell (1927, p. 267-268) suggested the following relationship between muscovite and phengite:



and stated that "it is understood that the iron in the phengite formula is ferrous and that magnesium may proxy for it in any amount, in fact, magnesium is more abundant than ferrous iron in nearly all modern analyses of phengitic micas." Thus the phengite formula could as well be written, $\text{K}(\text{Al}_{1.5}\text{Mg}_{0.5})(\text{Si}_{3.5}\text{Al}_{0.5})\text{O}_{10}(\text{OH})_2$.

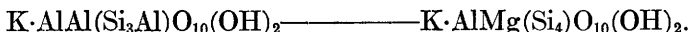
Muscovite and phengite have the same potassium content, indicating that replacements of higher valence cations by lower valence cations have induced the same amount of charge in the atomic layers. As phengite has only half as much aluminum proxying for silicon in the tetrahedral layers as muscovite, it has only half as much tetrahedral charge. The other half of the charge is provided by bivalent iron and magnesium proxying for aluminum in the octahedral layer. Thus, whereas in muscovite the charge is entirely tetrahedral, in phengite the charge is half tetrahedral and half octahedral.

The formula proposed by Winchell for phengite is a theoretical formula suggested to account for the high silica content and the presence of ferrous iron and magnesium in certain micas. In formulas derived from actual analyses the octahedral charge may be less than that required by the formula. For example, in the formula derived

from a phengite analysis reported by Jakob (1925), the octahedral charge amounts only to approximately one-third of the total charge: $X_{0.96}(Al_{1.50}Fe_{0.15}^{+3}Fe_{0.07}^{+2}Mg_{0.28})(Si_{3.40}Al_{0.60})O_{10}(OH)_2$. The formula also indicates that the specimen contains ferric as well as ferrous iron, but the dominant bivalent ion is magnesium.

From muscovite to phengite the trend is toward greater occupancy of the tetrahedral positions by silicon. If no compensatory change took place in the octahedral layers the end result of such a trend would be pyrophyllite. In order to maintain the charge and the potassium content constant, filling of the tetrahedral positions by silicon and the decrease of the charge in the tetrahedral layers must be accompanied by increasing proxying, ion-for-ion, of octahedral aluminum by bivalent ions with a consequently increasing charge in the octahedral layer. The limit of such a trend is complete occupancy of the tetrahedral positions by silicon and the filling of one of the two occupied octahedral positions by bivalent ions with the final location of the whole charge in the octahedral layer.

(Schaller (1950, p. 408) suggested that the composition of the high-silica sericites can be explained if they are considered members of such a trisilicic-tetrasilicic series,



In members of this series maintenance of a constant charge requires the increase in silicon content be accompanied by an equivalent increase in magnesium and by a decrease in aluminum content equivalent to the increase in silicon plus magnesium. In the end-members the charge is due, on the one hand, entirely to replacement in the tetrahedral layers, and on the other hand, entirely to replacement in the octahedral layer. In intermediate members of the series the charge is partly tetrahedral and partly octahedral.

In interpreting the composition of high-silica sericites Schaller calculated all Fe_2O_3 , Cr_2O_3 , and Mn_2O_3 reported in the analyses to equivalent Al_2O_3 , and all FeO and MnO as equivalent MgO . Hence Al_2O_3 and MgO were used by Schaller as collective terms but as other trivalent and bivalent constituents were present in only minor amounts, Al_2O_3 and MgO were the essential octahedral constituents in the specimens considered by him. This is also true of many other dioctahedral potassium micas but not of all, as is evident from the analyses and the derived formulas presented in tables 1 and 2. In some, constituents other than Al_2O_3 and MgO are the important octahedral constituents as, for example, ferrous and ferric iron in glauconite and celadonite, and vanadium in roscoelite.

The formulas in table 2 indicate that all the specimens they represent are members of a trisilicic-tetrasilicic series. In order to correlate

them, however, more generalized formulas than those suggested by Schaller are desirable, such as



in which the general symbols R^{+3} and R^{+2} are used to represent trivalent and bivalent ions in the octahedral group. X is used to represent the large cation balancing the total charge for the reason that in most of these micas, as in others, K is not entirely equivalent to the total charge; small amounts of Na or Ca are generally present. No generalization is needed for the tetrahedral group, made up as it is of varying proportions of Si and Al . From such generalized formulas any dioctahedral potassium mica can be placed in the series and thus can be correlated with other micas in this family, regardless of the constituent ions making up the octahedral group. Generalized forms of certain formulas selected from table 2 are listed in table 3. In these formulas the charge due to lower valence cations proxying for higher in the tetrahedral and octahedral groups are noted above these groups. The formulas in table 3 represent a complete and continuous series from the trisilicic to tetrasilicic end members. In the pink muscovite (formula 1) the entire charge is due to replacement of one-fourth of the tetravalent Si ions by trivalent aluminum ions in the tetrahedral group. In succeeding formulas there is a gradual shift in the seat of the charge, due to less and less replacement of Si by Al in the tetrahedral group and more and more replacement of trivalent by bivalent ions in the octahedral group, until in celadonite (formula 17) the tetrahedral group is entirely occupied by Si ions and bivalent ions occupy half of the two occupied octahedral positions.

It must be emphasized that in this series, proxying of octahedral trivalent ions by bivalent ions is ion-for-ion with no tendency toward filling the third unoccupied octahedral position. In the formulas shown in table 2 the maximum number of occupied octahedral positions is 2.03. In the tetrasilicic end-member, celadonite (formula 17), 2.01 octahedral positions are occupied.

The tetrahedral-octahedral charge relationships of the general formulas in table 3 are shown graphically in figure 13. The four columns of these histograms represent, from left to right, total charge, tetrahedral charge, octahedral charge, and potassium, in terms of equivalents of charge. The histograms show clearly that all proportions of tetrahedral charge to octahedral charge may be found among these micas. Progress from trisilicic to tetrasilicic is not stepwise but continuous, although formulas representing stages in the shift are shown above the histograms at appropriate places.

Alurgite was referred to by Stevens (1946, p. 106) as the tetrasilicic end-member of the $K \cdot AlAl(Si_3Al)O_{10}(OH)_2$ — $K \cdot AlMg(Si_4)O_{10}(OH)_2$

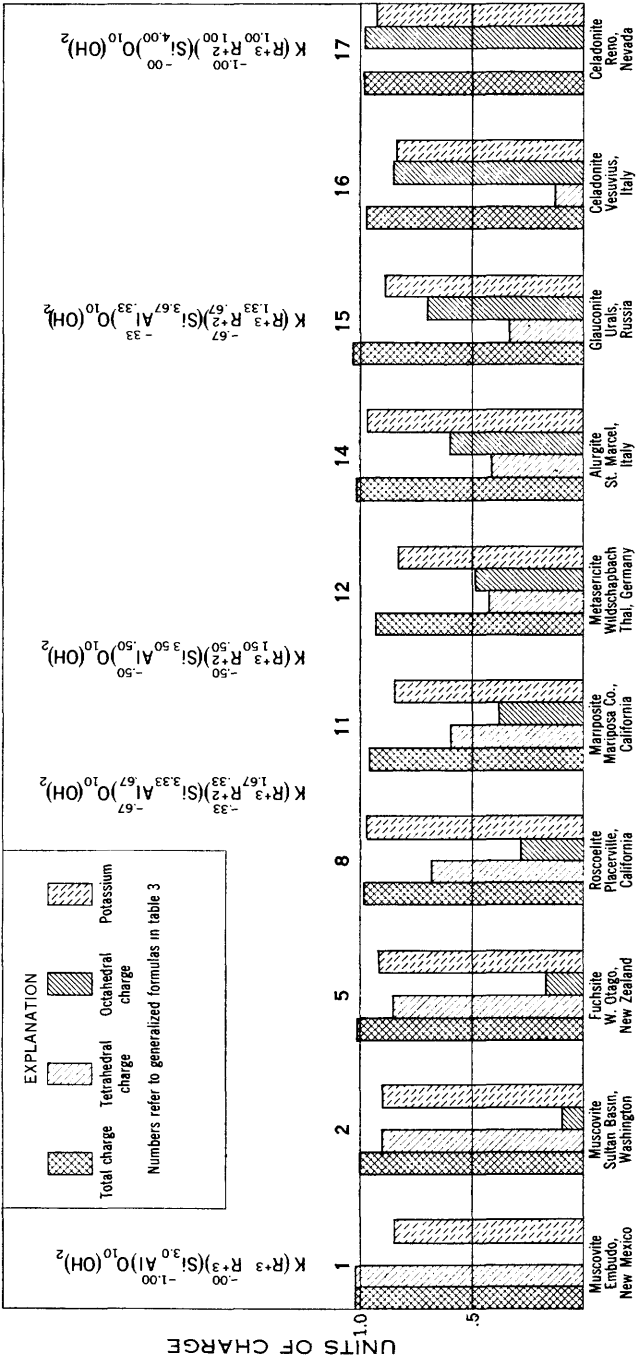


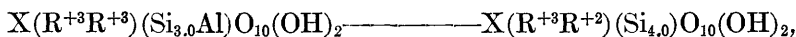
FIGURE 13.—Tetrahedral-octahedral charge relationship in selected dioctahedral potassium micas.

series. However, with aluminum occupying 0.4 positions in the tetrahedral group it is very little beyond the middle of the series. On the other hand, it is the most tetrasilicic dioctahedral potassium mica known in which aluminum is the predominant trivalent octahedral ion. Leucophyllite, referred to by Schaller (1950, p. 407) as the tetrasilicic end-member of the above series, cannot be considered from available analyses a potassium mica as it has only 3.39 percent K_2O , only one-third of the required amount.

The tetrasilicic end of the general series is represented, therefore, only by the high-iron dioctahedral potassium micas glauconite and celadonite. These materials are commonly considered hydrous micas, rather than true micas. Hendricks and Ross (1941) did not clearly differentiate between micas and hydrous micas among the glauconites and celadonites, as they were considering the origin and chemical composition of these materials. However, among the analyses cited by them are some whose potassium-water relation is that of true micas, not hydrous micas. One of these, a celadonite (table 2, formula 17) is the tetrasilicic end-member of the general series. It has no tetrahedral aluminum; the tetrahedral group is completely occupied by silicon. This specimen has been examined by Dr. H. S. Yoder of the Geophysical Laboratory (oral communication) and found to be a 1M mica.

The high-iron dioctahedral micas dominate the tetrasilicic end of the general series, but they do not (insofar as is known) extend beyond the middle of the series toward the trisilicic end. The failure of the aluminum members to extend far beyond the middle of the series toward the tetrasilicic end has already been noted. Thus, neither aluminum nor iron as the dominant trivalent octahedral ion forms a complete trisilicic-tetrasilicic series.

The trisilicic-tetrasilicic general series,



affords a system for classifying and correlating the dioctahedral potassium micas on the basis of their tetrahedral-octahedral charge relationship. By this system the relationship between commonly known micas of this family, muscovite and phengite, and those having unusual or complex composition (table 2) the roscoelites (formulas 4 and 8), fuchsite—chromium muscovite—(formula 5), mariposite (formula 11), or alurgite (formula 14) is clarified and the reverse-charge relationships between muscovites and phengites and glauconites and celadonites is apparent. Thus the great variety of composition found in this family of minerals, the result of isomorphous replacements, is simplified and reduced to the single factor of the charge induced on the structural layers by the replacement.

HYDROUS MICAS

The same method can be used for the classification and correlation of hydrous micas. However, hydrous micas commonly occur in nature very intimately mixed or even interlayered with other minerals and are very difficult, often impossible, to purify. Other materials called hydrous micas may in reality be mixtures of others minerals—as of muscovite and montmorillonite. Such a mixture could be very similar in composition to some materials that have been called illite. It would yield a rational formula and have a comparable tetrahedral-octahedral charge relationship. The effect of montmorillonite in a mixture with a mica or hydrous mica is to cause a decrease in the total charge, the tetrahedral charge, and the potassium content, as montmorillonite is characterized by low total charge (less than half that of muscovite), little or no tetrahedral charge, and very little potassium. It also tends to increase the octahedral charge, as most of the charge in montmorillonite is octahedral. The greater the proportion of montmorillonite, the greater these effects would be. Other minerals, such as chlorite, kaolinite, and montmorillonite, often found intermixed or interlayered with fine-grained micas or hydrous micas, may or may not yield a mixture the composition of which can be expressed by rational formulas. The fact that a rational formula can be derived from an analysis does not guarantee that there is only one mineral present. This, however, does not invalidate the use of this method for classifying and correlating materials that are called hydrous micas.

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TABLE 1. *Analyses of dioctahedral potassium micas*

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
SiO ₂	44.80	46.77	46.80	44.81	46.35	48.10	49.16	45.17	50.05	50.79	55.35	52.58	49.75	53.22	49.53	55.30	55.61
Al ₂ O ₃	37.72	34.75	35.84	18.42	29.69	32.30	30.51	11.54	30.11	26.53	25.62	23.56	7.52	21.19	5.84	10.90	17.19
FeO	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
MgO	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
CaO	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Na ₂ O	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
K ₂ O	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Cr ₂ O ₃	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
MnO ₂	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
MnO	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
V ₂ O ₅	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
TiO ₂	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
H ₂ O	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
F	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Total	100.18	100.04	100.11	99.73	100.31	100.14	100.07	99.86	100.05	100.17	99.73	100.13	99.91	99.99	99.54	99.61	100.27

18 percent quartz deducted.

2 Includes 0.38 Li₂O, 0.82 Rb₂O, 0.09 Cs₂O.

3 Includes 0.13 BaO.

4 Includes 0.15 BaO.

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17. Celadonite, 23 mi. E. of Reno, Nev. R. C. Wells, 1937, U. S. Geol. Survey Bull. 878, p. 102.

TABLE 2.—*Formulas for dioctahedral potassium micas.*

1. Muscovite (pink)---	$X_{1.01}(Al_{1.93}Fe_{.03}^{+3}Mn_{.01}^{+3}Li_{.05})(Si_{2.98}Al_{.02})O_{10}(OH)_2$ K _{.85}	+0.01	-1.02
2. Muscovite-----	$X_{.99}(Al_{1.84}Fe_{.04}^{+3}Fe_{.04}^{+2}Mg_{.09})(Si_{3.11}Al_{.89})O_{10}(OH)_2$ K _{.90}	-.10	-.89
3. Sericite-----	$X_{.96}(Al_{1.93}Fe_{.01}^{+3}Mg_{.06})(Si_{3.12}Al_{.88})O_{10}(OH)_2$ K _{.85}	-.07	-.88
4. Roscoelite-----	$X_{.93}(Al_{.73}V_{1.16}^{+3}Fe_{.09}^{+2}Mg_{.02})(Si_{3.18}Al_{.82})O_{10}(OH)_2$ K _{.75}	-.11	-.82
5. Fuchsite (Chromian muscovite)-----	$X_{1.02}(Al_{1.62}Fe_{.01}^{+3}Cr_{.25}^{+3}Fe_{.05}^{+2}Mg_{.19})(Si_{3.15}Al_{.85})O_{10}(OH)_2$ K _{.91}	-.17	-.85
6. Gilbertite-----	$X_{1.01}(Al_{1.76}Fe_{.18}^{+2}Mg_{.06})(Si_{3.22}Al_{.78})O_{10}(OH)_2$ K _{.85}	-.23	-.78
7. Sericite-----	$X_{1.01}(Al_{1.70}Fe_{.08}^{+3}Mg_{.22})(Si_{3.28}Al_{.72})O_{10}(OH)_2$ K _{.93}	-.29	-.72
8. Roscoelite-----	$X_{.98}(Al_{.31}V_{1.41}^{+3}Fe_{.10}^{+2}Mg_{.18})(Si_{3.31}Al_{.69})O_{10}(OH)_2$ K _{.97}	-.29	-.69
9. Sericite (phengite)--	$X_{1.07}(Al_{1.63}Fe_{.07}^{+3}Fe_{.02}^{+2}Mg_{.24})(Si_{3.29}Al_{.71})O_{10}(OH)_2$ K _{.82}	-.36	-.71
10. Phengite-----	$X_{.96}(Al_{1.50}Fe_{.15}^{+3}Fe_{.07}^{+2}Mg_{.28})(Si_{3.40}Al_{.60})O_{10}(OH)_2$ K _{.83}	-.36	-.60
11. Mariposite (green)--	$X_{.95}(Al_{1.59}Fe_{.03}^{+3}Cr_{.01}^{+3}Fe_{.06}^{+2}Mg_{.32})(Si_{3.41}Al_{.59})O_{10}(OH)_2$ K _{.85}	-.36	-.59
12. Metasericite-----	$X_{.92}(Al_{1.45}Fe_{.33}^{+3}Mg_{.25})(Si_{3.57}Al_{.43})O_{10}(OH)_2$ K _{.82}	-.49	-.43
13. Glauconite-----	$X_{.88}(Al_{.29}Fe_{1.22}^{+3}Fe_{.14}^{+2}Mg_{.35})(Si_{3.62}Al_{.38})O_{10}(OH)_2$ K _{.84}	-.50	-.38
14. Alurgite-----	$X_{1.01}(Al_{1.28}Fe_{.06}^{+3}Mn_{.04}^{+3}Fe_{.01}^{+2}Mg_{.61})(Si_{3.59}Al_{.41})O_{10}(OH)_2$ K _{.96}	-.60	-.41
15. Glauconite-----	$X_{1.03}(Al_{.18}Fe_{1.12}^{+3}Fe_{.37}^{+2}Mg_{.32})(Si_{3.67}Al_{.33})O_{10}(OH)_2$ K _{.88}	-.70	-.33
16. Celadonite-----	$X_{.97}(Al_{.78}Fe_{.37}^{+3}Fe_{.20}^{+2}Mg_{.65})(Si_{3.88}Al_{.12})O_{10}(OH)_2$ K _{.84}	-.85	-.12
17. Celadonite-----	$K_{.98}(Al_{.07}Fe_{.93}^{+3}Fe_{.24}^{+2}Mg_{.77})(Si_{4.00})O_{10}(OH)_2$ K _{.92}	-.98	-.00

TABLE 3.—Generalized formulas for selected dioctahedral potassium micas

1. Muscovite (pink)-----	$X_{1.00}(R_{1.97}^{+3}R_{.05}^{+2})(Si_{2.98}Al_{1.02})O_{10}(OH)_2$
2. Muscovite-----	$X_{.99}(R_{1.88}^{+3}R_{.13}^{+2})(Si_{3.11}Al_{.89})O_{10}(OH)_2$
5. Fuchsite (Chromian muscovite)-----	$X_{1.02}(R_{1.78}^{+3}R_{.24}^{+2})(Si_{3.15}Al_{.85})O_{10}(OH)_2$
8. Roscoelite-----	$X_{.98}(R_{1.72}^{+3}R_{.28}^{+2})(Si_{3.31}Al_{.69})O_{10}(OH)_2$
11. Mariposite-----	$X_{.95}(R_{1.63}^{+3}R_{.37}^{+2})(Si_{3.41}Al_{.59})O_{10}(OH)_2$
12. Metasericite-----	$X_{.92}(R_{1.46}^{+3}R_{.54}^{+2})(Si_{3.57}Al_{.43})O_{10}(OH)_2$
14. Alurgite-----	$X_{1.01}(R_{1.38}^{+3}R_{.62}^{+2})(Si_{3.59}Al_{.41})O_{10}(OH)_2$
15. Glauconite-----	$X_{1.03}(R_{1.30}^{+3}R_{.69}^{+2})(Si_{3.67}Al_{.33})O_{10}(OH)_2$
16. Celadonite-----	$X_{.97}(R_{1.15}^{+3}R_{.85}^{+2})(Si_{3.88}Al_{.12})O_{10}(OH)_2$
17. Celadonite-----	$X_{.98}(R_{1.00}^{+3}R_{1.01}^{+2})(Si_{4.00})O_{10}(OH)_2$

