

An interpretation of the composition of high-silica sericites

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Abstract: The high-silica sericites generally contain a corresponding appreciable quantity of a divalent element, usually magnesium, and their chemical composition is interpreted as being intermediate between that of muscovite, $KAl_2(AlSi_3)O_{10}(OH)_2$, and that of the equivalent high-silica mica leucophyllite, $KA1Mg(Si_4)O_{10}(OH)_2$. The series muscovite-leucophyllite includes the named micas phengite (high-silica sericite), mariposite, and alurgite. Selected analyses are plotted and fall on a corresponding straight compositional variation line from muscovite to leucophyllite. It is shown diagrammatically that the analysed sample of mariposite probably contained about 8 % of quartz. It is recommended that the only species names in this series to be retained are those of the end members muscovite and leucophyllite. Their indices of refraction and specific gravities are very similar.

Introduction.

ANALYSES of many sericites agree closely with analyses of many muscovites, the difference in the two minerals being essentially in the state of physical aggregation. Other sericites contain a larger amount of silica than is contained in muscovite and have in part been termed phengite. These high-silica sericites generally contain more divalent elements (chiefly magnesium and iron) than those sericites whose silica content is about the same as that of nonsericitic muscovite. The reported high-silica content of those sericites with only very small amounts of divalent elements is probably due to admixed quartz. It is the purpose of this paper to emphasize this content of divalent elements in high-silica sericites as the basis for an interpretation of their chemical composition.

Mica formulae.

An interpretation of the composition of any mica must be based on the now well-known structural formulae of the micas, which can be generalized as $X.Y_{2-3}.Z_4.O_{10}(OH)_2$. X represents the large cations (Na, Ca, K, Ba, &c.), with an ionic radius of about 1.0 to 1.4; Y represents cations (Al, Fe, Mg, Li, &c.) in octahedral co-ordination, with an intermediate ionic radius of about 0.6 to 0.9; and Z represents cations (Si, Al) in tetrahedral co-ordination, with a smaller ionic radius of about

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0.4 to 0.6. Anions (O, F, &c.), with a large ionic radius of about 1.3 and larger, may substitute for hydroxyl. In the Y position a dioctahedral¹ formula has two cations with one vacant octahedral position,² and a trioctahedral formula has three cations with no vacant position. In the Z position are four cations with tetrahedral co-ordination. These are Si and Al and they may be either a tetrasilicic group (Si₄), a trisilicic group (AlSi₃), a disilicic group (Al₂Si₂), or a monosilicic group (Al₃Si).² Some examples of the end-member mica formulae, on this basis, are shown in the tabulation below.

Series (from one end-member formula to another end-member formula) exist in the micas either wholly within one of these groups or between two of them. Hence in a mica formula calculated from an analysis, the number of cations of an element need not be a whole number as the cations are in the end-member formulae listed below.

Some examples of end-member mica formulae.

Tetrasilicic:

Polyolithionite	K.LiLiAlSi ₄ O ₁₀ F ₂
Taeniolite	K.LiMgMgSi ₄ O ₁₀ F ₂
Celadonite*	K.Fe ^o Mg ^o Si ₄ O ₁₀ (OH) ₂
Leucophyllite†	K.AlMg ^o Si ₄ O ₁₀ (OH) ₂

Trisilicic:

Muscovite	K.AlAl ^o AlSi ₃ O ₁₀ (OH) ₂
Phlogopite	K.MgMgMgAlSi ₃ O ₁₀ (OH) ₂
Siderophyllite	K.Fe ^o Fe ^o Fe ^o AlSi ₃ O ₁₀ (OH) ₂
Barium muscovite‡	Ba.AlMg ^o AlSi ₃ O ₁₀ (OH) ₂

* For this formula of celadonite see Hendricks and Ross (1941).

† This formula was referred to as alurgite by Stevens (1946), p. 106, on my suggestion, but as the analyses of leucophyllite are closer to the magnesium end-member of the muscovite-leucophyllite series (see fig. 1), this name seems preferable and is so used. Starkl (1883), p. 657, coined the name leucophyllite as a rock name for a phyllite with a glistening white colour and extraordinary fine platy structure. However, apparently he considered the predominant component of the phyllite as one mineral, for he says: 'Die Härte des Minerals ist 1-2.' The sample analysed was '... möglichst reine quarzfreie Substanz...' Dana (1892), p. 616, uses the term leucophyllite as a mineral name for a variety of muscovite. Should future investigations show that Starkl's analysis was made on non-homogeneous material (mixture of sericite, talc, &c.) and the term leucophyllite become untenable as a mineral species name, then alurgite would be the name most applicable to what is here called leucophyllite.

‡ Not known as a mineral. A calcium muscovite would have a similar formula. The formula given for barium muscovite requires 31.06 % BaO and 8.17 % MgO. An analysis of a barium-containing muscovite from Franklin, New Jersey, described

¹ Following the nomenclature proposed by Stevens (1946).

² The writer has found it most useful, in studying various group formulae, to have in the formulae a symbol indicating a vacant position, and suggests a small open circle, raised halfway up in the line of type, such as ^o. This symbol is here used.

Disilicic:

Margarite	Ca. $AlAl^{\circ}.Al_2Si_2.O_{10}(OH)_2$
Magnesium margarite§	Ca. $MgMgMg.Al_2Si_2.O_{10}(OH)_2$
Ephesite;	Na. $AlAlLi.Al_2Si_2.O_{10}(OH)_2$
Willcoxite¶	Na. $AlMgMg.Al_2Si_2.O_{10}(OH)_2$

Monosilicic:

Xanthophyllite	Ca. $AlMgMg.Al_3Si.O_{10}(OH)_2$
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by Bauer and Berman (1933), shows 9.89 % BaO (and 6.33 % K₂O, 1.51 % Na₂O, and 0.36 % CaO) with 1.55 % MgO (and 1.84 % ZnO and 0.62 % MnO), and hence the mica analysed is of intermediate composition. Oellacherite is similar in composition.

§ Not known as a mineral. Brandisite and seybertite are intermediate members of the magnesium margarite-xanthophyllite (*monosilicic*) series.

¶ A new analysis of ephesite from Postmasburg, South Africa, made in the U.S. Geological Survey, has shown that the previous determinations of Li₂O are much too low. The mineral contains nearly 4 % of Li₂O and forms a series with margarite. A paper on the margarite-ephesite series, by W. T. Schaller and M. K. Carron of the U.S. Geological Survey is in preparation.

¶ A neglected member of the disilicic micas. Genth's analysis (Genth, 1873) shows a little iron and potassium. The atomic ratios for the analysis are: Z = 4.00 (1.98 Si and 2.02 Al); Y = 2.95 (0.99 Al, 0.07 Fe^{'''}, 0.14 Fe^{''}, and 1.75 Mg; X = 1.06 (0.85 Na and 0.21 K); (OH) = 1.66.

The total charges of the cations in these formulae are 22. They may be distributed as follows:

	Large cation.	Octahedral group.	Tetrahedral group.
Tetrasilicic	1	5	16
	2	4	16
Trisilicic	1	6	15
	2	5	15
Disilicic	1	7	14
	2	6	14
Monosilicic	1	8	13
	2	7	13

Hence in going from trisilicic muscovite with 15 charges in the tetrahedral group to a tetrasilicic formula, such as that of leucophyllite, with 16 charges in the tetrahedral group, as long as the large cation (like potassium) remains monovalent, the charges in the octahedral group must be reduced from 6 to 5. This is readily accomplished by substituting one bivalent magnesium (or one ferrous iron) for one trivalent aluminium. As a high-silica mica must be in part tetrasilicic, there must also be present the corresponding amount of a bivalent element, to achieve compensation in the total cation charges. That is why high-silica sericites must contain the equivalent amount of magnesium or some other bivalent element of the proper ionic radius.

The composition of the high-silica sericites can be explained as being

members of a series from trisilicic muscovite, $\text{KAlAl}(\text{AlSi}_3)\text{O}_{10}(\text{OH})_2$, to tetrasilicic leucophyllite, $\text{KAlMg}(\text{Si}_4)\text{O}_{10}(\text{OH})_2$. The Si_4 in the leucophyllite formula accounts for the high silica and the Mg (or Fe^{II}) accounts for the necessary presence of divalent elements.

We shall now see how the available data on the high-silica sericites agree with this explanation of their composition.

Discussion of chemical analyses.

Seven analyses are discussed. Except for the two analyses with highest percentages of SiO_2 and of MgO (namely, those of alurgite and leucophyllite), only analyses made in the U.S. Geological Survey are considered. Analyses of five varieties of pure sericite from Amelia, Virginia, are given by Glass (1935). As the analyses of two pairs of varieties are very close, their results are averaged:

1. Average of the analyses of the purple sericites, varieties 3 and 4 (Glass). Amelia, Virginia. R. E. Stevens, analyst.

2. Average of the analyses of the water-green sericite, variety 1, and of the maize-yellow sericite, variety 2. Amelia, Virginia. J. G. Fairchild, analyst.

3. White, claylike sericite from Melones, Calaveras County, California. U.S. Geol. Survey Chem. Lab. Record no. D-124 (no. 2). $2V = 0^\circ$ to $10^\circ r$. J. J. Fahey, analyst.

4. Marguerite-yellow to chalcedony-yellow sericite, variety 5. Amelia, Virginia. R. E. Stevens, analyst.

Additional analyses of high-silica sericites with several per cent. of MgO, which are very close to the analyses of the sericites from Amelia, are listed by Dana (1892) on pp. 618-619.

5. Green mariposite from Mariposa County, California, analysed by Hillebrand (1900). A second analysis of white mariposite from the same locality in which the ferrous and ferric oxides were not separately determined is very similar and hence is not used in this paper. As here shown, the green sample analysed contained about 8 % of quartz. According to Knopf (1929), p. 44, 'The mariposite-ankerite masses are generally traversed by a network of veinlets, consisting of coarse milky-white quartz.' He notes, on p. 38, the presence of rutile, accounting for the TiO_2 , and states that mariposite is 'practically uniaxial', with a maximum value of 36° for $2E$.

5a. The same analysis, after deducting 8 % of quartz and recalculating to 100 %. The method of arriving at the amount of quartz present is explained farther on.

6. Alurgite from San Marcel, Italy, analysed by Penfield (1893). This is the only one of the seven selected samples which does not have the fine scaly or fibrous texture with a silky or waxy luster commonly ascribed to sericite. Through the courtesy of Prof. Horace Winchell of Yale, some fragments of Penfield's type specimen (Brush collection no. 5523) were obtained. These fragments have β or γ indices of refraction about 1.597 to 1.600. Some scales were uniaxial, others had a $2V$ of 26° . Meixner (1939) obtained similar results.

7. Leucophyllite from Anna-Capelle, Austria, described and analysed by Starkl (1883). Analysis no. 49 on p. 619 of Dana (1892).

These seven analyses are reproduced in the tabulation below, to which are added the calculated percentage compositions of muscovite (first column) and of leucophyllite (last column) according to the end-member formulae given above. As will

be noted, there is a regular increase in the percentages of SiO_2 and of MgO (including FeO and MnO) with a corresponding decrease in the percentages of Al_2O_3 (including Fe_2O_3 , Cr_2O_3 , and Mn_2O_3).

Tabulation of selected analyses of high-silica sericites.

	M Muscovite end-member	Sericites			Mariposite			7 Leucophyllite Austria	L Leucophyllite end-member	
		1	2	3	4	5	5a			6
SiO_2	45.26	46.81	47.64	49.37	49.16	55.35	51.39	53.22	57.11	60.57
Al_2O_3	38.40	35.97	32.07	29.21	30.81	25.62	27.81	21.19	19.80	12.85
Fe_2O_3	—	—	—	1.54	—	0.63	0.68	1.22	2.99	—
Mn_2O_3	—	—	—	—	—	—	—	0.87	—	—
Cr_2O_3	—	—	—	—	—	0.18	0.20	—	—	—
FeO	—	0.25	1.16	—	1.43	0.92	1.00	—	—	—
MnO	—	—	0.17	—	—	—	—	0.18	—	—
MgO	—	0.59	1.32	2.77	2.22	3.25	3.53	6.02	8.85	10.17
K_2O	11.82	10.16	9.05	9.72	10.90	9.29	10.08	11.20	3.39	11.87
Na_2O	—	0.64	0.09	0.14	0.48	0.12	0.13	0.34	1.42	—
H_2O	—	0.53	2.60	0.75	0.15	—	—	—	—	—
H_2O^+	4.52	5.03	5.56	6.88	4.73	4.52	4.91	5.75	6.30	4.54
etc.	—	0.30	0.12	0.10	0.19	0.25	0.27	—	0.49	—
	100.00	100.28	99.78	100.57	100.07	100.13	100.00	99.99	100.35	100.00

Plotting analyses quoted.

The percentage values for SiO_2 , Al_2O_3 , and MgO are plotted for each analysis on the rectangular diagram shown in fig. 1 as the three pairs of SiO_2 against Al_2O_3 , SiO_2 against MgO , and Al_2O_3 against MgO . The three resulting points for each analysis should coincide and should lie on the straight line joining the similar points for end-member muscovite, $\text{KAlAl}(\text{AlSi}_3\text{O}_{10})(\text{OH})_2(\text{M})$, and for end-member leucophyllite, $\text{KAlMg}(\text{Si}_4\text{O}_{10})(\text{OH})_2(\text{L})$. As the values used for M and L are based on SiO_2 , Al_2O_3 , and MgO , it becomes necessary to recalculate the analyses considered on the same basis. Therefore the percentages of Fe_2O_3 , Cr_2O_3 , and Mn_2O_3 are calculated to their equivalent Al_2O_3 ; FeO and MnO to MgO ; and Na_2O and CaO to K_2O . The resultant values are then recalculated to a basis of 100%. The percentages so obtained, which are plotted in fig. 1, are as follows:

Recalculated percentages of SiO_2 , Al_2O_3 , and MgO .

Analyses	1	2	3	4	5	5a	6	7
SiO_2	46.48	48.01	49.33	49.26	55.60	51.74	53.57	56.92
Al_2O_3	35.72	32.30	30.17	30.87	26.26	28.54	22.69	21.63
MgO	0.73	2.08	2.77	3.03	3.79	4.12	6.16	8.82

In plotting the values for Al_2O_3 and MgO , the ordinates for MgO are used, whereas in plotting the values for SiO_2 and MgO , the abscissae for MgO are used.

The placements of the three points for each analysis gives some idea

as to the purity of the sample as they should coincide and fall on the line M-L, as they practically do for all analyses except those of mariposite (no. 5) and of leucophyllite (no. 7). The large circle necessary to

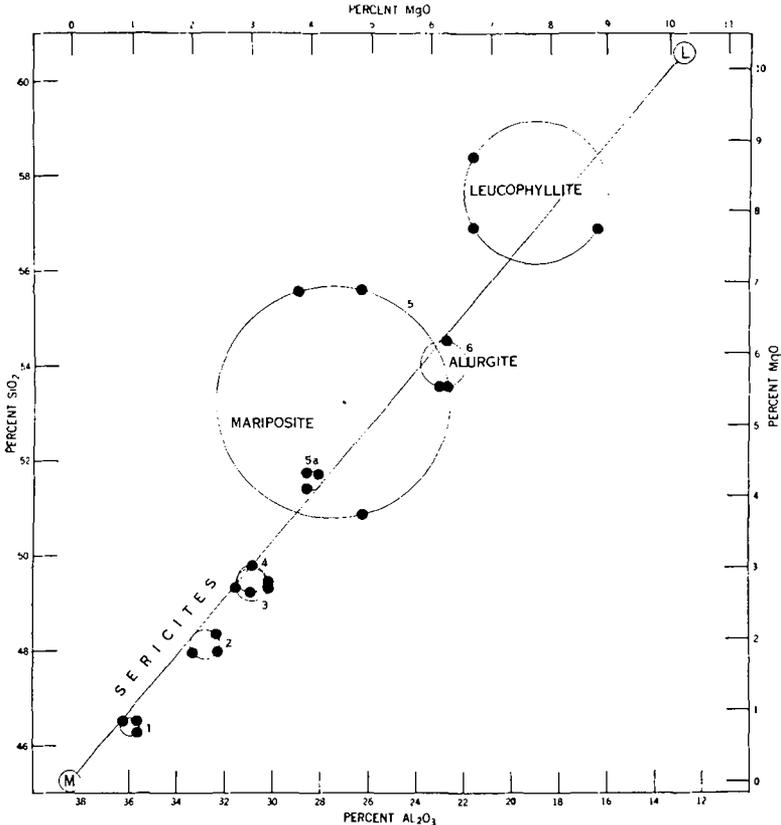


FIG. 1. Percentages of SiO_2 against Al_2O_3 , SiO_2 against MgO , and Al_2O_3 against MgO , plotted as solid dots. The three dots for each analysis are then joined by a circle. For no. 4 two of the dots coincide. All three dots for each analysis should coincide and lie on the straight line joining the positions for end-member muscovite (M) and for end-member leucophyllite (L) if the sample analysed was homogeneous. The size of the circumscribed circle gives an estimate of the size of the impurities.

circumscribe the three dots for the analysis of mariposite (no. 5) indicates that the sample analysed was impure. As shown in fig. 2 the sample contained about 8 % of quartz. When this is deducted and the analysis recalculated to 100 % the resulting three dots (no. 5a) fall nearly on the line M-L and the resulting circle is very small.

The method of arriving at the conclusion that about 8 % of quartz was present in the sample of mariposite analysed by Hillebrand is as follows. His analysis was recalculated on the basis of deducting respectively 2, 4, 6, 8, 9, 10, 12, and 14 % of quartz, the three pairs of percentages plotted, and circles drawn through the three dots. The resultant circles indicate the extent of the impurities. If the circles are large, it is evident that the analysis is incorrect or that the sample analysed was impure.

By plotting the three points for the analysis of mariposite, after deducting the amounts of quartz stated, the circles first decrease in size, as shown in fig. 2, the smallest circle representing the results of deducting 8 % of quartz. If 9 % of quartz is deducted, the circles become larger again, indicating that 8 % represents closely the amount of quartz which must have been present in the sample analysed.

The three plotted points for alurgite (no. 6) fall closely on the M-L line and the resulting circle is small. Alurgite is closer to the leucophyllite end of the series than mariposite. Stevens (1946), p. 112, calculated the atomic ratios of Penfield's analysis to be: Z = 4.00 (3.59 Si and 0.41 Al); Y = 2.00 (1.27 Al, 0.06 Fe^{III}, 0.05 Mn^{III}, 0.01 Mn^{II}, and 0.61 Mg); X = 1.00 (0.96 K and 0.04 Na); (OH) = 2.59. Alurgite is 59 % tetrasilic leucophyllite and 41 % trisilic muscovite.

For leucophyllite two analyses are given by Starkl (1883) with almost identical percentages of MgO but with about 4 % difference in the SiO₂ and nearly as much for Al₂O₃. These are analyses 49 and 50 as listed by Dana (1892) on p. 619. The presence of hydrated iron oxide, staining some flakes yellowish-brown, was noted by Starkl. Both analyses report nearly 3 % Fe₂O₃ with no determination of FeO. Plotting the values for the analysis with the lowest SiO₂ (52.81 %) resulted in such a large circle that evidently either the analysis is faulty or the sample was impure. Consequently this analysis is not further considered, though both the high SiO₂ and the high MgO show that its placement on the M-L line is near the leucophyllite end.

The second analysis by Starkl (no. 7 in fig. 1) with 57.11 % SiO₂ was made on material freed from quartz as far as possible. Applying the procedure used for interpreting the analysis of mariposite as made on a mixture of sericite and quartz did not yield similar results. The resultant circles, after deducting respectively 2 and 4 % of quartz, became larger instead of smaller, indicating that no quartz was present in the sample analysed. It is believed that the percentage of MgO reported is probably too high. The very low content of alkali with corresponding high

water, if correct, may be the reason that the three dots cannot be brought closer together. The low alkali content indicates a considerable trend

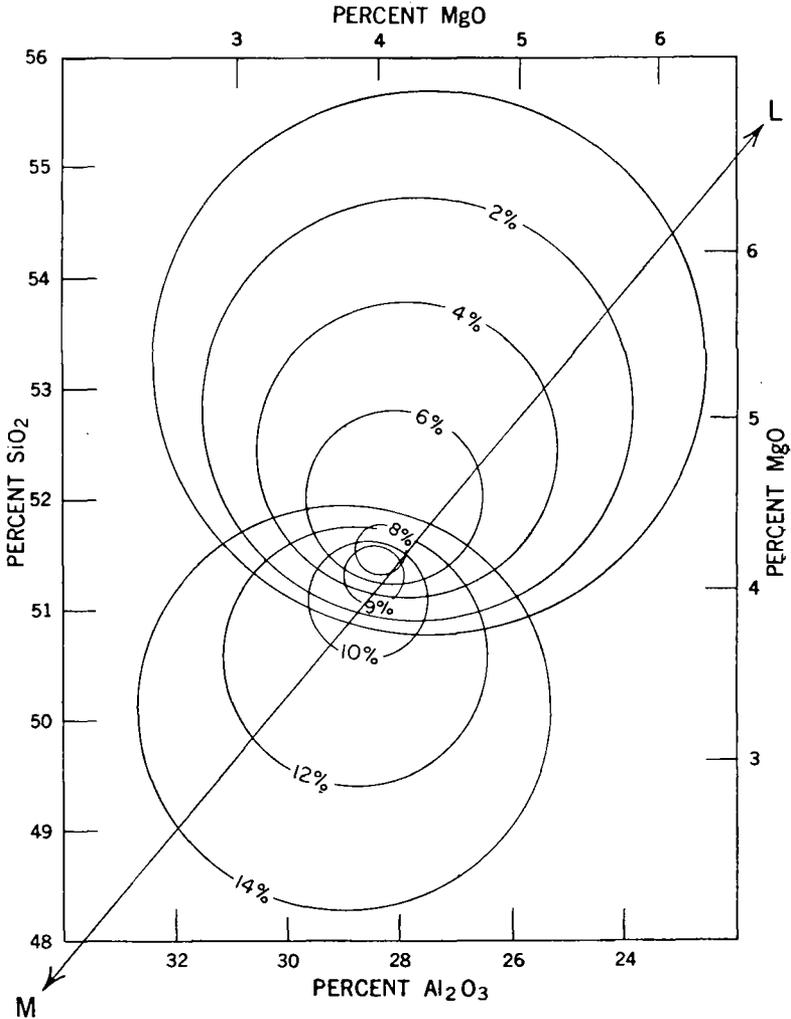


Fig. 2. The percentages of SiO_2 , Al_2O_3 , and MgO plotted as described for fig. 1 for the analysis of mariposite, after deducting respectively 2, 4, 6, 8, 9, 10, 12, and 14 % of quartz. The smallest circle joining each group of three dots is the one for 8 % of quartz; the largest circle represents the original analysis.

towards a hydromica. That the high-silica sericites show a definite trend towards the hydromicas is not confirmed by the other analyses plotted.

Both mariposite and alurgite are close to the middle of the *M-L* line and the two minerals are similar in composition, as previously noted by Schaller (1916), who suggested their essential identity. The so-called alurgite from Cajón Pass, California, described by Webb (1939), is not alurgite and is not simply a member of the muscovite-leucophyllite series. It is not a high-silica mica. It contains only 46.30 % SiO_2 , whereas if it were a member of the muscovite-leucophyllite series, the 4.92 % of $\text{MgO} + \text{MnO}$ present would require about 53 % of SiO_2 . Meixner (1939), like Webb (1939), regards the red colour of the Italian alurgite as an essential property of alurgite. The writer prefers to regard the colour as an unessential property, depending on the presence of a relatively very small amount of a chromatic element. He would define alurgite as a part of the muscovite-leucophyllite series with necessary high-silica and the presence of an equivalent amount of divalent elements, usually magnesium. However, neither of the two names alurgite and mariposite deserves species rank. Both should be discarded, though for local interest the name mariposite will probably persist, as this green mica is so characteristically of widespread occurrence in the ankeritized rock derived from serpentine in the Mother Lode region of California. Alurgite is nearer the leucophyllite end of the muscovite-leucophyllite series and should be referred to as a variety of leucophyllite, should this name be adopted for the high-silica end-member of the series, with the formula $\text{KAlMgSi}_4\text{O}_{10}(\text{OH})_2$.

It has not been possible to correlate systematically the variations in indices of refraction and in specific gravity with the variations in chemical composition for the members of the series. The indices of refraction of alurgite (the closest member of the series to leucophyllite for which data are available) are essentially those of muscovite. It has been stated that high-silica sericites have a very small axial angle, but so do some muscovites (see Axelrod and Grimaldi, 1949). Both the indices of refraction and specific gravity of muscovite and of leucophyllite are probably very close. The presence of small amounts of chromatic elements, such as Cr in mariposite and Mn in alurgite, probably causes a greater difference in the indices of refraction than that existing between muscovite and leucophyllite. Knopf (1929) gives the values of 1.60, 1.61, and 1.63 for the gamma index for different samples of mariposite.

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