OPTICAL AND CHEMICAL STUDIES OF MUSCOVITE

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Abstract

Twenty-two samples of muscovite were analyzed chemically and their optical constants determined. From these results, the chemical compositions of the end members of the muscovite system were determined and their relation to the optical constants correlated. The chemical formulas for the members of the muscovite system which correspond most accurately with the chemical composition are potassium muscovite ($H_4K_2Al_6Si_6O_{24}$), phengite ($H_6K_2(Fe, Mg)_2Al_4Si_6O_{24}$), (new formula), and ferric iron muscovite ($H_4K_2Fe_2'''Al_4Si_6O_{24}$). It was necessary to adopt this new formula for phengite, which contains one more molecule of H_2O and FeO or MgO, and one less molecule of SiO_2 than the formula now being used, in order to get satisfactory correlation with the results of the chemical analyses. The ratio of R_2O (K_2O and Na_2O) to SiO_2 remains almost constant in all three members at 1 to 6 instead of varying between 1 to 5 and 1 to 8 as suggested by various investigators. When the ferric iron muscovite of the system increases, the refractive index rises. The optic angle decreases with an increase in the amount of phengite, however, the optic angle also decreases when small amounts of phengite are present with large amounts of ferric iron muscovite.

Introduction

This paper reports the results of an investigation of the optical properties and chemical composition of the muscovite system. The literature contains many studies bringing out the relationship of the optics of muscovites to their chemistry, but there is lack of complete agreement on the formulas to be assigned to the component end members. A modified formula for phengite is suggested here.

If the composition of muscovite is expressed on the basis of the constituent molecules instead of on the basis of the oxides, the variation in optical properties is more easily understood. This method has been employed effectively by others and is followed here, leading to three end member molecules. Our interest centers on the formulas to be assigned to these molecules.

Present Theories

As early as 1879 Tschermak (10) pointed out that a number of muscovite samples contained more silica than is required by the general formula given to this mica. These high silica micas were designated "phengite" and often have a small optic angle. Along this same line Winchell (13)

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has published a summary of many analyses of muscovite and has concluded that the muscovite system has three end members, one of which accounts for the high silica. He gives the following formulas for this system:

Muscovite $H_4K_2Al_6Si_6O_{24}$
Phengite $H_4K_2(Fe, Mg)Al_4Si_7O_{24}$
Fe''' Muscovite $H_4K_2Fe_2'''Al_4Si_6O_{24}$

The phengite which contains $7SiO_2$ instead of $6SiO_2$ accounts for the high silica. He considers one silicon atom and one ferrous iron or magnesium atom proxies for two aluminum atoms. After arriving at satisfactory end members, the optical properties were correlated with the percentage end member composition. He found that ferric iron decidedly increased the refractive index and birefringence, and that ferrous iron and magnesium decreased the optic angle. Kunitz (8) also found that the iron in micas increased the refractive index and birefringence.

In 1925 Hallimond (5), in working with muscovite found that the K_2O to SiO_2 ratio remained fairly constant at 1 to 6 and that when this ratio did vary it was due to variations in the K_2O, with the SiO_2 remaining constant. Many analyses were also found that contained high percentages of water and fluorine. These were accounted for by considering both mutually replaceable, and to be in the lattice structure as $Al(OH)_2$ and $Al(F)_2$. He also indicated that ferrous iron and magnesium can replace the aluminum to form phengite, thus giving a system with two end members.

In reviewing the literature there were found several points of dispute that have not been settled, and in order to correlate the optical properties of muscovite accurately with its chemical composition, the members of the muscovite system must be defined. Hundreds of analyses have been published which are widely different and are from material of unknown purity, thus making it extremely difficult to come to a satisfactory conclusion. With this in view, the results reported in this paper were carried out on samples of high quality and were further checked for their purity by methods recently devised.

Micas Studied

The micas used in this study were selected from the mineral collections of the Geology Department of the University of Wisconsin and the supply of Ward's Natural Science Establishment. A list of the micas studied, the locality from which they were collected, and the source from which they were obtained are given in Table 1.

TABLE 1. LOCALITY OF OCCURRENCE AND SOURCE OF SUPPLY OF THE MUSCOVITES STUDIED

Muscovite	Locality of Occurrence	Source of Supply
1 M	South Dakota	University of Wisconsin
2 "	Haywood, North Carolina	" " "
3 "	Phenocite Mine, Villa Rio, Piracicoba, Brazil	" " "
4 "	Branchville, Connecticut	" " "
5 "	Kokomo, Colorado	" " "
6 "	Brazil	" " "
7 "	Big Ridge Mica Mine, Haywood, North Carolina	" " "
8 "	Mitchell County, North Carolina	" " "
9 "	India	Wards Nat. Sci. Estb.
10 "	Grafton, New Hampshire	" " " "
11 "	Delaware County, Pennsylvania	" " " "
12 "	Montville, New Jersey	" " " "
13 "	Auburn, Maine	" " " "
15 "	Haddam Neck, Connecticut	" " " "
16 "	Branchville, Connecticut	" " " "
17 "	Central Australia	" " " "
18 "	Bamle, Norway	" " " "
19 "	Burke Falls, Ontario	" " " "
21 "	Dobrova, Corinthia	" " " "
23 "	Easton, Pennsylvania	" " " "
24 "	Mitchell County, North Carolina	" " " "
25 "	Keystone, South Dakota	" " " "

In a study of this nature, it is extremely important to have samples free of inclusions of other minerals and of unweathered material. To obtain these, samples of muscovite were selected from high grade pegmatite material. Special care was exercised in selecting only that part of the specimen that was of the greatest purity.

METHODS OF ANALYSES

The samples of muscovite were ground to pass a 100 mesh sieve and then separated by means of the specific gravity method described by Volk (11) to remove any impurities in the form of inclusions. The material was now ready for chemical analysis. Any free iron oxide remaining was first removed and determined by treating with H_2S and acidifying according to Drosdoff and Truog (2). The iron thus extracted was subtracted from the total ferric iron to get the amount of ferric iron actually in the silicate molecule. These methods of selection and purification made it possible to obtain total analyses representative of extremely pure muscovite.

All chemical analyses, except those of sodium and potassium, were carried out according to the standard methods outlined by Hillebrand and Lundell (6). The sodium was determined by the sodium zinc uranyl acetate method described by Chapman (1) and the potassium determined by the sodium cobalti-nitrite method according to Volk and Truog (12).

The determinations of the refractive indices were made according to the double variation method of Emmons (4).

Results and Discussion

The results from the optical analyses of the muscovites are given in Table 2 and those of the chemical analyses in Table 3.

Table 2. Optical Data of Muscovites

Muscovite	Refractive indices			Observed optic angle	α-γ	α-β	β-γ	F-D Dispersion
	α	β	γ	Degrees				
1 M	1.5943	1.5894	1.5602	44	.0341	.0049	.0292	.0070
2 "	1.6035	1.5994	1.5677	38	.0358	.0041	.0317	.0070
3 "	1.5988	1.5940	1.5637	39.0	.0351	.0048	.0303	.0064
4 "	1.6010	1.5955	1.5649	42.5	.0361	.0055	.0306	.0040
5 "	1.6066	1.6019	1.5592	40	.0474	.0047	.0427	.0057
6 "	1.6034	1.5980	1.5544	41	.0490	.0054	.0436	.0059
7 "	1.6042	1.5995	1.5625	37	.0417	.0047	.0370	.0051
8 "	1.5987	1.5946	1.5577	39	.0410	.0041	.0369	.0058
9 "	1.5951	1.5902	1.5457	39.8	.0494	.0049	.0445	.0087
10 "	1.5985	1.5927	1.5511	41.8	.0474	.0058	.0416	.0066
11 "	1.6113	1.6062	1.5591	37.5	.0522	.0051	.0471	.0072
12 "	1.6029	1.5968	1.5569	43	.0460	.0061	.0399	.0060
13 "	1.5910	1.5855	1.5574	45	.0336	.0055	.0281	.0063
15 "	1.5937	1.5884	1.5587	45	.0350	.0053	.0297	.0067
16 "	1.6026	1.5973	1.5616	41	.0410	.0053	.0357	.0042
17 "	1.5989	1.5945	1.5738	44.3	.0251	.0044	.0207	.0096
18 "	1.6100	1.6043	1.5669	41	.0431	.0057	.0374	.0062
19 "	1.6091	1.6045	1.5711	43.0	.0380	.0046	.0334	.0090
21 "	1.5943	1.5899	1.5512	38	.0431	.0044	.0387	.0069
23 "	1.6114	1.6065	1.5647	37.5	.0467	.0049	.0418	.0071
24 "	1.6110	1.6056	1.5667	38	.0443	.0054	.0389	.0085
25 "	1.5973	1.5914	1.5603	43	.0370	.0059	.0311	.0059

All indices given are $\pm$.0003.

TABLE 3. CHEMICAL COMPOSITION OF THE MUSCOVITES

Mus-covite	SiO_2 %	Al_2O_3 %	K_2O %	Na_2O %	MgO %	FeO %	Fe_2O_3 %	CaO %	TiO_2 %	Loss on ignition %	Total
1 M	44.25	34.67	10.74	0.54	1.07	1.47	0.00	0.23	trace	6.08	99.05
2 "	44.80	34.50	9.48	1.47	2.40	0.65	0.64	0.00	0.73	5.53	100.20
3 "	45.07	33.95	9.98	1.26	0.51	3.68	0.00	0.17	0.25	5.81	100.68
4 "	42.96	34.24	9.97	1.09	0.63	3.39	0.00	0.09	0.09	6.88	99.34
5 "	44.78	33.80	9.29	1.58	1.10	0.75	1.80	0.34	0.30	6.20	99.94
6 "	43.97	35.58	9.57	1.39	1.82	0.96	0.45	0.00	0.35	6.61	100.70
7 "	42.60	33.60	9.28	1.59	1.57	1.46	0.70	0.80	0.89	7.82	100.31
8 "	42.90	33.21	9.65	1.70	1.37	1.45	0.00	0.00	0.58	9.73	100.59
9 "	42.35	36.50	9.70	1.30	1.60	1.40	0.37	0.50	0.58	6.70	101.00
10 "	44.25	36.98	9.30	1.53	1.50	1.40	0.01	0.31	0.22	5.10	100.65
11 "	44.15	35.35	10.20	1.01	0.89	0.86	2.15	0.28	0.44	5.30	100.65
12 "	43.50	35.41	10.04	1.28	0.80	0.89	0.84	0.31	0.00	6.74	99.81
13 "	44.75	36.51	10.30	0.82	0.41	1.41	0.00	0.00	0.00	5.20	99.40
15 "	44.65	36.63	10.30	0.90	0.61	0.85	0.00	0.20	0.00	5.92	100.06
16 "	43.43	33.92	10.90	0.54	0.75	4.47	0.00	0.00	trace	5.48	99.49
17 "	44.68	34.98	10.35	1.40	0.60	1.04	0.00	0.00	0.39	5.56	99.00
18 "	43.37	33.19	10.17	1.03	1.36	1.00	1.95	0.00	0.33	7.74	100.14
19 "	44.77	33.26	10.58	0.78	1.21	1.34	1.82	0.26	0.18	6.02	100.22
21 "	45.45	34.41	10.44	1.40	3.11	0.40	0.12	0.17	0.31	4.77	100.58
23 "	45.10	33.22	9.98	1.15	0.94	1.21	2.75	0.00	0.25	6.27	100.87
24 "	44.09	33.01	10.30	0.95	1.00	0.66	2.00	0.00	0.10	8.86	100.97
25 "	44.15	34.96	9.81	1.21	1.00	1.32	0.00	0.14	0.11	7.53	100.23

THE CHEMICAL CLASSIFICATION OF THE MUSCOVITE SYSTEM

The results of the chemical analysis of the muscovites studied have been evaluated according to the methods used by Winchell (15) and Hallimond (5) to determine the likely formulas for the end members of this system. The compositions for Winchell's suggested end members were based upon the amounts of FeO, MgO, Fe_2O_3, and K_2O determined. These were calculated to the following formulas, $H_4K_2Al_6Si_6O_{24}$ (muscovite), $H_4K_2(Fe''Mg)Al_4Si_7O_{24}$ (phengite), and $H_4K_2Fe_2'''Al_4Si_6O_{24}$. If these are the correct end members of the muscovite system, the results of the calculations should show no consistent lack or abundance of any one element. Table 4 presents the percentage molecular composition of the muscovites here reported. These results show a consistent lack of silica to fulfill the requirements of these end members. Because of this consistent deficiency of silica, a new formula—$H_6K_2(Fe \text{ or } Mg)_2Al_4Si_6O_{24}$—is suggested for the phengite molecule. In this formula there are two RO groups replacing one R_2O_3 group, thus eliminating the necessity of using silicon for this purpose. The water is increased from two to three molecules which corresponds fairly well to the amounts of water found by analysis. Table 4 also presents the percentage composition of the end members, two of which are the same as those used by Winchell, and the third or new member containing $3H_2O$, $2FeO$ or MgO, and $6SiO_2$ instead of $2H_2O$, FeO or MgO, and $7SiO_2$. The results show a very good correlation between these members and the chemical composition.

Hallimond (5) computes all of his data on the basis that $SiO_2 = 600$, thus showing the various ratios found in the analyses. Table 5 presents the data calculated according to this system and sets forth some very interesting relations. The K_2O to SiO_2 ratio remains almost constant at 1 to 6. In only one case out of 22 does the $RO + R_2O_3$ to SiO_2 ratio go below 1 to 3 and that one case is 1 to 2.99. This indicates that it is not necessary for silicon to proxy for aluminum and eliminates any necessity of accounting for high silica in any of the muscovites analyzed. By computing the analyses to the two suggested formulas of Hallimond; namely, muscovite, $K_2O \cdot 3R_2O_3 \cdot 6SiO_2 \cdot 2H_2O$, and phengite, $K_2O \cdot RO \cdot 2R_2O_3 \cdot 6SiO_2 \cdot 2H_2O$, the silica checked within experimental error in 18 cases out of 22, but the aluminum was generally in excess of 1.5 per cent. The aluminum checked within 1.5 per cent excess or deficiency in 18 out of 22 cases when 2RO were substituted for one R_2O_3 as is suggested for the new phengite molecule.

TABLE 4. THE MOLECULAR COMPOSITION OF MUSCOVITE BASED ON THE PERCENTAGE OF POTASSIUM MUSCOVITE, PHENGITE, AND FERRIC IRON MUSCOVITE MOLECULES

Muscovite	Per cent molecular composition						Per cent of SiO_2 and Al_2O_3 not required or lacking to fulfill the formulas by			
	by Winchell's end members			by using new phengite formula			Winchell		New phengite	
	$H_4K_2Al_6Si_6O_{24}$	$H_4K_2(Fe, Mg)$-$Al_4Si_7O_{24}$ (phengite)	$H_4K_2Fe_2'''$-$Al_4Si_6O_{24}$	$H_4K_2Al_6Si_6O_{24}$	H_6K_2-$(Fe''Mg)_2$-$Al_4Si_6O_{24}$	$H_4K_2Fe_2'''$-$Al_4Si_6O_{24}$	SiO_2	Al_2O_3	SiO_2	Al_2O_3
1 M	63.2	36.8	0	80.5	19.5	0	−2.7	+2.0	0	−0.4
2 "	43.3	55.5	3.2	69.8	27.0	3.2	−3.1	+3.7	+0.8	+0.2
3 "	48.5	51.5	0	74.4	25.6	0	−3.3	+2.5	+0.4	−0.8
4 "	48.0	52.0	0	74.0	26.0	0	−4.1	+3.6	−0.3	+0.5
5 "	61.5	30.4	9.5	75.5	15.4	9.1	−1.7	+1.0	+0.4	−0.6
6 "	50.8	47.0	2.2	74.4	23.4	2.2	−4.0	+3.7	−0.5	+0.7
7 "	48.9	47.5	3.6	72.6	23.8	3.6	−5.0	+2.0	−1.4	−0.9
8 "	58.2	41.8	0	76.5	23.5	0	−7.3	−0.8	−3.9	−4.0
9 "	50.2	48.0	1.8	74.0	24.2	1.8	−5.4	+4.6	−2.0	+1.5
10 "	55.0	45.0	0	77.5	22.5	0	−3.6	+4.8	0	+2.1
11 "	61.7	27.4	10.9	75.4	13.7	10.9	−2.3	+2.3	−0.2	+0.5
12 "	70.0	25.6	4.4	82.0	13.6	4.4	−7.0	+0.4	−1.8	−1.1
13 "	75.0	25.0	0	87.5	12.5	0	−1.2	+2.1	+0.4	+0.5
15 "	78.2	21.8	0	89.0	11.0	0	−1.2	+1.7	+0.2	+0.2
16 "	35.0	65.0	0	70.0	30.0	0	−9.0	+4.2	−1.2	0
17 "	78.0	22.0	0	89.0	11.0	0	−4.4	−2.7	−2.7	−4.2
18 "	52.0	38.2	9.8	71.0	19.2	9.8	−3.9	+1.3	−1.1	0
19 "	51.5	39.0	9.5	71.0	19.5	9.5	−2.8	+1.2	0	−1.3
21 "	38.0	62.0	0	69.0	31.0	0	−7.2	+2.2	−2.2	−2.0
23 "	53.6	32.4	14.0	69.8	16.2	14.0	−2.0	+1.0	−1.0	+0.4
24 "	50.5	39.5	10.0	70.2	19.8	10.0	−3.6	+1.3	−1.2	−0.7
25 "	65.0	35.0	0	82.4	17.6	0	−2.7	+1.6	−0.1	−0.5

TABLE 5. THE MOLECULAR EQUIVALENTS OF THE OXIDES, OF THE MUSCOVITES STUDIED, COMPUTED ON THE BASIS OF $SiO_2+TiO_2=600$

No.	Al_2O_3	Fe_2O_3	FeO	MgO	$RO+R_2O_3$	R_2O	H_2O
1	277	—	17	21	315	100	276
2	267	3.2	7.5	45	323	98	243
3	266	—	41	10	317	99	257
4	280	—	39	13	332	101	320
5	266	9	8.3	21.8	305	99	275
6	284	2.3	11.0	36.7	334	101	300
7	275	—	16.8	31.0	323	103	370
8	270	—	15.8	30	316	108	448
9	301	1.9	16.4	32.2	351	105	313
10	295	—	15.9	29.3	340	100	230
11	280	11.0	9.7	17.8	318	101	248
12	288	4.4	10.3	16.6	319	105	310
13	291	—	16.6	8.5	316	99	235
15	291	—	9.6	12.0	313	99	264
16	276	—	51.5	15.3	337	103	252
17	274	—	11.6	11.6	299	105	247
18	269	10.1	11.4	27.8	318	103	355
19	261	9.1	15.0	24.0	309	100	267
21	267	—	4.3	61.0	332	105	210
23	269	13.8	13.5	18.5	315	99	276
24	265	10.2	7.5	32.9	316	102	400
25	280	—	15.1	20.2	315	101	341

The new formula for the phengite member presented in this paper appears to be in full accord with the chemical composition of the muscovites studied. A reduction of the silica from $7SiO_2$, as given by Winchell, to $6SiO_2$, as used in the new formula, accounts for the deficiency of silica noted when $7SiO_2$ was used. This change in silica content leaves the phengite with only 23 oxygens and in order to increase the oxygen content to 24 the water has been changed from two to three molecules. There is sufficient water recorded in the analyses to account for this change in water. It is not uncommon to find many muscovites with high water content and as suggested by Hallimond, it may be accounted for in the crystal lattice in groups such as $Al(OH)_2$ and AlO, or an interchange between them. There is a possibility that the extra hydroxyl radical may be combined with the ferrous iron as in $Fe(OH)_2$. There is another change in the new phengite molecule formula which is also substantiated by the group of muscovites studied in this work. This change is the replacement of $1R_2O_3$ group by 2RO groups instead of an exchange of 1RO for $1R_2O_3$, as suggested in the old formula. Thus we have built up a new formula for the phengite molecule which is not radically different in form from the other two members of the muscovite system and which coincides satisfactorily with the chemical composition of the muscovites reported in this paper.

Relation of Optical Properties to Chemical Composition

In determining the relation of the optical properties to the chemical composition of muscovite, the end members selected are those suggested in an earlier part of this paper; namely, potassium muscovite

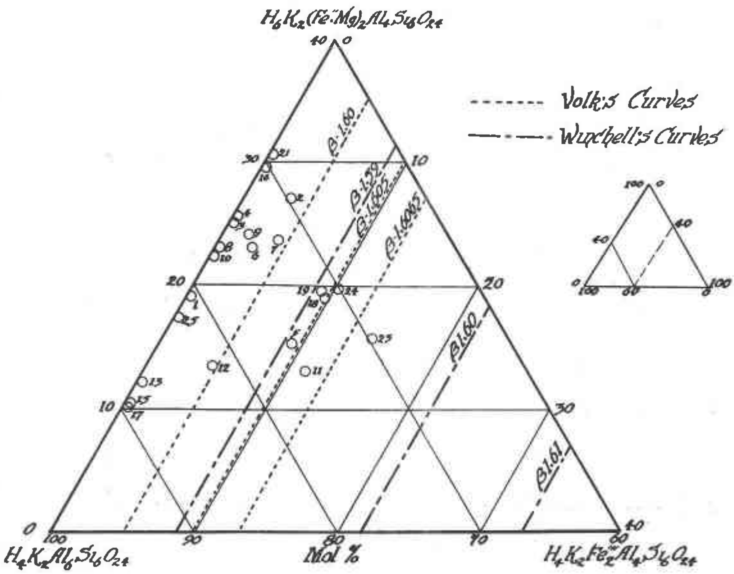

FIG. 1. Variations in the composition and refractive index in the muscovite system.

($H_4K_2Al_6Si_6O_{24}$), phengite ($H_6K_2(Fe''Mg)_2Al_4Si_6O_{24}$), and ferric iron muscovite ($H_4K_2Fe_2'''Al_4Si_6O_{24}$). The end member compositions were then plotted on a three component diagram as shown in Fig. 1. This is the same type of diagram used by Winchell (14), and the lines drawn through it indicate the values of β determined in this study and those reported by Winchell (14). The results show that the values of β increase with increasing amounts of the ferric iron muscovite, which is in accord with the findings of Winchell.

The effect of the phengite content of the muscovite system on the optic angle is shown in Fig. 2. When the phengite increases, the optic angle decreases, but when there are considerable amounts of ferric iron present along with small amounts of phengite, the same effect is registered on the optic angle. To further check this effect of ferric iron, some analyses of muscovites by Jakob (7), Smirnoff (9) and Eckermann (3) were plotted on the same graph. These show the same effect.

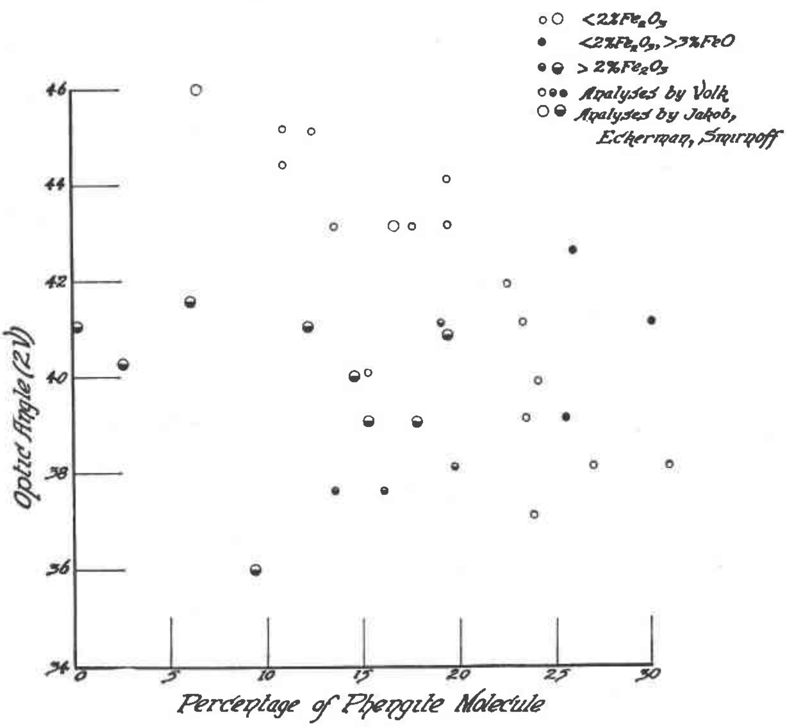

FIG. 2. The effect of the phengite molecule on the optic angle.

A study of dispersion and birefringence did not show any correlation with the chemical composition, but this may be due to the limited number of samples used in this investigation.

In order that the true relations may be expressed between the chemical composition and the optical properties of a mineral, it is necessary to use results from materials of high purity. The formulas expressing the actual form of the minerals present must also be used.

The relations expressed in the diagrams between the optical properties and the chemical compositions are only general, and are not so accurately established as is desired. This, however, is because there was an insufficient number of samples used and the range was incomplete even though the extremes in optical properties were covered. The differences between the values of β as given by Winchell and those reported here may be explained as being due to the use of purer samples in the latter case, which was made possible by the use of improved methods of purification. These methods removed considerable amounts of free ferric iron which has no effect on the refractive index. After the removal of the free ferric iron any effect on the refractive index by iron would have to be caused by that which is in the silicate molecule.

The effectiveness of the phengite molecule in reducing the optic angle is much greater than that recorded by Winchell. This is due to the nature of the new phengite molecule which has 2RO groups in place of 1RO group and thus reduces the phengite one-half in the muscovite system. It appears that muscovite specimens of high quality must be used if more accurate relations between the optical properties and chemical composition are to be established.

Summary

The purpose of this investigation was to obtain more information on the optical properties and chemical classification of the muscovite system. Twenty-two samples of muscovite, first purified by means of heavy liquids, and then the remaining free iron oxide removed by treating with H_2S and acidifying, were used in this study. These muscovite samples were then analyzed chemically and their optical constants determined. A summary of the results follows:

The chemical formulas for the members of the muscovite system which correspond most accurately with the chemical composition are: potassium muscovite ($H_4K_2Al_6Si_6O_{24}$), phengite ($H_6K_2(Fe,Mg)_2Al_4Si_6O_{24}$), and ferric iron muscovite ($H_4K_2Fe_2'''Al_4Si_6O_{24}$). It was necessary to adopt this new formula for phengite in order to get satisfactory correlation with the results of chemical analyses.

When the ferric iron muscovite content of the system increases, the refractive index rises.

The optic angle decreases with an increase in the amount of phengite. The optic angle also decreases when small amounts of phengite are present with large amounts of ferric iron muscovite.

No correlation has been established between the birefringence or dispersion and the chemical composition of the muscovites analyzed.

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