MORPHOLOGY AND CRYSTAL CHEMISTRY OF 1:1 LAYER LATTICE SILICATES*

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Abstract

Sixty-four chemical analyses of sheet structure silicates with a 1:1 type layer lattice have been evaluated in order to demonstrate certain interrelationships between chemistry, structure and morphology within and between the following mineral groups: kaolin, serpentine and "other 1:1" layer lattice silicates (amesite, cronstedtite and ferrous and ferric chamosite). The analyses of five synthetic 1:1 compounds and seven representative chlorites are also included and discussed.

The morphological characteristics of the minerals relate directly to the nature and amount of cation substitution in tetrahedral and octaheral sheets, and to the amount of H_2O+. The former is evaluated in terms of a morphological index, M, which gives a measure of the amount of misfit of the two sheets within the layer. The effect of the latter is shown in ternary composition diagrams.

Structural formulas are computed on the basis of a lattice of 18 oxygens but using the premise that oxygen and (OH) allocation between the two sheets will be proportioned in accord with the cation distribution. The allocation of cations, in turn, is based on the assumption that substitutions or additions of Fe''' and Al in the octahedral sheet will equal substitutions of these ions for silicon in the tetrahedral sheet.

The kaolin, serpentine and "other 1:1" groups differ from each other chiefly with respect to the amount of cation substitution and therefore the amount of misfit of the two sheets in the 1:1 layer. In terms of the morphological index, serpentines range from $+73.53$ to $+60.09$, other 1:1 from $+35.35$ to -60.09 and kaolins from -72.11 to -84.13.

Within the serpentine and kaolin groups cation substitutions play an important role but the amount of hydrogen provides the most important distinction between platy and tubular varieties. Analyses of chrysotile and halloysite ($2H_2O$) contain significantly more H_2O+ than those of their platy counterparts. Although the position of the "excess" hydrogen and oxygen ions in or on the lattice is not known, the effect of these elements is apparently to weaken interlayer bonds to the extent that misfit of the sheets produces tubes or curved laths rather than plates.

The evaluation of the chlorite analyses yields results similar to those obtained for amesite, chamosite and cronstedtite in that cation substitution provides for much less misfit than in kaolins or serpentines, and H_2O+ is nearly equal to or less than that to be expected in the O and (OH) of the properly proportioned structural formulas.

Introduction

Research during the past eight years on morphological details of kaolin, serpentine and related minerals has led to an evaluation of the relationship of morphology to chemical composition in those compounds which combine a "gibbsite" or "brucite" sheet with a Si-O sheet to give a so-called 1:1 layer structure. Meanwhile studies of others on crystal structure, density and other characteristics of the minerals have pro-

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vided additional information as to the relationship of the minerals to each other and to other members of the sheet structure silicate group.

In the present study the minerals of major interest are those of the kaolin and serpentine groups but, in order to obtain a more complete picture of overall chemical-morphological relationships, amesite, cronstedtite, chamosite and several synthetic compounds having the 1:1 structure are included. In addition, the relationship of the chlorites to these minerals is considered.

Summary of Morphological Data

Amesite and cronstedtite both occur in small but megascopic crystals, amesite in the form of hexagonal plates, cronstedtite in hexagonal pyramids and diverging groups having perfect basal cleavage.

Chamosite normally is massive or oolitic. Deudon (1955) points out that in electron micrographs platy particles are commonly irregular but sometimes show hexagonal corners.

In the kaolin group, nacrite, dickite and kaolinite are platy and show poor to well-developed hexagonal outlines depending on the source and type of material and the manner of preparation of the material for observation in the electron microscope. Flat laths of well-crystallized kaolinite with hexagonal terminations are not uncommon, particularly from areas where hydrothermal solutions have led to the formation of clay minerals.

At the other extreme in the kaolin group are the irregular nodules and spherical particles considered typical of allophane (Davis, et al., 1950, p. 9; Sudo and Takahashi, 1956). The latter authors have shown that there may be a morphological and chemical transition between amorphous spherules of allophane and the tubes of halloysite ($4H_2O$). Replicas of fractured surfaces of samples of the latter (Bates and Comer, 1955) demonstrate that tubes are the characteristic crystal form of this clay mineral, and show that the cross sections of the cylinders are circular or elliptical in outline.

The occurrence of tubes with polygonal outlines (Bates, 1955) plus evidence of the existence of lath-shaped particles in a mixture of equidimensional kaolinite plates and halloysite tubes (Brindley and Comer, 1956), led to a more critical re-evaluation of hundreds of electron micrographs of halloysite, and to a detailed study of certain clay samples using replicas of fractured surfaces in order to minimize disturbance of the particles during sample preparation. The results of this study were presented at the Sixth National Clay Conference and are published in the proceedings thereof (Bates and Comer, 1959) but may be briefly summarized as follows. Replicas of fractured surfaces of halloysite

($2H_2O$) clay from the Fox deposit, Utah County, Utah, and of a halloy-
site ($2H_2O$)-kaolinite clay from the Raddatz deposit near Eureka,
Utah, show that most of the halloysite particles are lath-shaped and
possess crystallographic terminations and angles which indicate a higher
degree of crystallinity than that commonly attributed to halloysite
($4H_2O$) tubes.* Figure 1, an electron micrograph of a fracture surface
of the Fox Clay, illustrates these features. Electron micrographs of dis-
persions of this clay show that the laths have a tendency to bend along
lines approximately parallel to the long axis of the particle thus giving
rise to tubes with the polygonal outlines of the type referred to above.
This evidence indicates that there is a complete morphological transition
in the kaolin group from the well-crystallized hexagonal plates of
kaolinite to elongate plates to laths with crystallographic terminations
to curved laths to tubes of halloysite ($4H_2O$). If the halloysite-allo-
phane transition suggested by Davis, *et al.* and discussed in more detail
by Sudo and Takahashi is correct, the morphological transition may be
said to extend all the way from the amorphous allophane to the well-
ordered kaolinite.
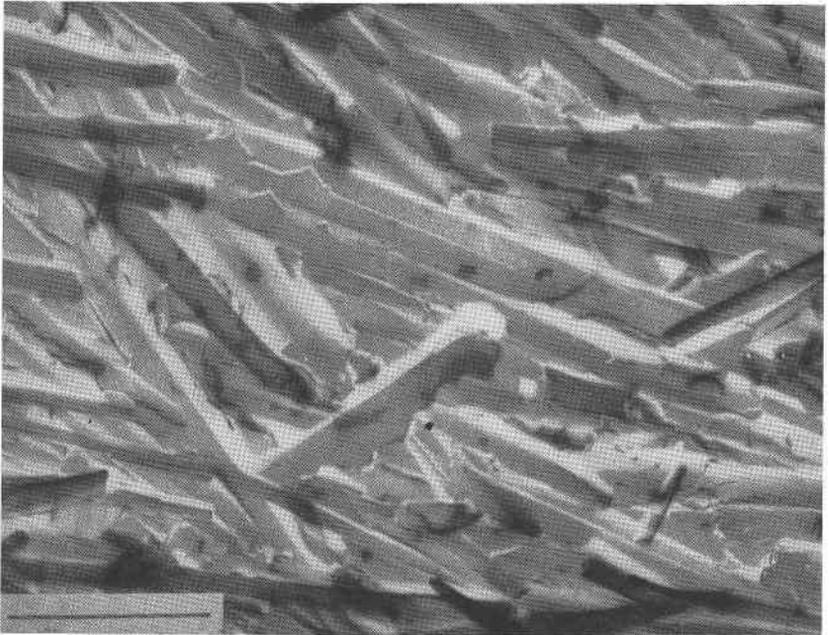
FIG. 1. Halloysite ($2H_2O$), Fox deposit, Utah County, Utah. Replica of fracture surface
showing laths with angular projections along edges. Scale represents one micron.

* This was observed and illustrated by Alexander, Faust, Hendricks, Insley and Mc-
Murdie (1943).

The analogy between the kaolin and serpentine groups is striking. Fibrous serpentine, or chrysotile, ranges morphologically from tubes of remarkable uniformity in inner and outer diameter and very high length /width ratio to elongate splintery laths many of which are associated with and genetically related to tubes. These give way to the broader curved laths and irregular sheets characteristic of antigorite and other structural varieties of the platy serpentine group. The rectified wave or corrugated structure postulated by Onsager (1952) and elaborated on by Zussman (1954) would appear to be morphologically intermediate between the curved laths of chrysotile and the well-developed plates of irregular outline but uniform thickness characteristic of the serpentine from Kennack Cove described by Midgely (1951) and later by Zussman, Brindley, and Comer (1957, p. 142), and referred to as lizardite by Whittaker and Zussman (1956). This material may represent the closest approach made by a natural serpentine mineral to the well-formed hexagonal crystals of synthetic serpentine such as those made by Tu (1950), Yoder (1952), and Roy and Roy (1954) by introducing Al into the serpentine structure.

Detailed morphological features of chrysotile tubes and halloysite laths and tubes, and the relationship of morphology to bulk density determinations have been dealt with in the paper referred to previously (Bates and Comer, 1959).

CHEMICAL DATA

Analyses Employed

Table I lists the analyses evaluated and their sources. In most cases the analyses used were those from recent literature wherein x-ray and other data were also reported for the samples. Chrysotile analysis #5 and platy serpentine analysis #5 are exceptions and represent the average of 29 and 14 analyses, respectively, labeled as chrysotile and antigorite in earlier literature. Dickite analyses D-1 to D-6, kaolinites K-5 to K-14, and halloysites H-2 to H-13 inclusive are taken from the reports by Ross and Kerr (1930, 1934) and the numbers used herein are those assigned in these papers.

The following synthetic compositions studied and described by Roy and Roy (1954) are also discussed and are designated in the figures by the numbers given below:

1) $Si_4Mg_6O_{10}(OH)_8$
2) $Si_4Ni_6O_{10}(OH)_8$
3) $Ge_4Mg_6O_{10}(OH)_8$
4) $Ge_4Ni_6O_{10}(OH)_8$
5) $(Si_3Al)(Mg_5Al)O_{10}(OH)_8$

In addition, characteristic compositions of penninite and corundo-

phyllite are used to help represent certain limits of chlorite composition. The formulas and the symbols used to represent them are:

P   Penninite $(Si_{3.2}Al_{.8})(Al_{.8}Mg_{1.6}Fe''_{3.6})O_{10}(OH)_8$
Co   Corundophyllite $(Si_{2.6}Al_{1.4})(Al_{1.4}Mg_{2.8}Fe''_{1.8})O_{10}(OH)_8$
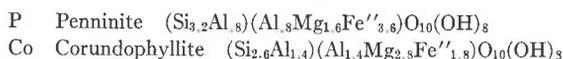
Calculation of Structural Formulas

In calculating from the analyses the structural formulas which will be used as a basis for pointing out some of the chemical-morphological relationships it is appreciated that experimental evidence is insufficient to permit a decision as to which of many possible arrangements of ions is most nearly correct. The procedure followed here was chosen primarily on the basis of simplicity and consistency with current concepts.

In making the assignments of cations to specific lattice sites the basic premise is somewhat different from that usually employed. Because of the layer structure of this particular group of minerals and because of the

TABLE 1. CHEMICAL ANALYSES

	C-1	C-2	C-3	C-4	C-5	C-6	C-7	C-8	P-1(M)	P-2(M)	P-3(M)	P-4(M)
SiO_2	41.70	42.50	41.80	41.88	41.84	42.02	41.33	41.24	41.52	42.40	41.80	42.02
Al_2O_3	.24	.32	.11	.19	.36	.52	.80	.06	.57	.74	.19	.49
Fe_2O_3	.30	.50	.68	.81	.52	.19	1.29	2.40	.36	.48	.93	1.50
MgO	42.85	42.50	42.82	41.38	40.83	41.44	41.39	38.43	42.80	42.70	42.67	42.05
FeO	.05	.01	.05	.05	2.00	.11	.08	2.16	.06	.043	.06	.057
MnO	.02	.02	.04	.05	—	.03	.04	.14	.04	.02	.04	.024
NiO	.00	.00	.00	.00	—	—	—	—	.00	.00	.00	.002
H_2O+	14.23	13.46	14.04	14.22	14.02	14.04	13.66	13.42	14.36	13.52	13.88	13.74
TiO_2	.03	.021	.05	.036	—	none	.02	trace	.06	.070	.10	.073
Cr_2O_3	.005	.004	.003	.005	—	—	—	—	.01	.008	.003	.001
CaO	.05	.04	.10	.47	—	none	trace	—	.11	.01	.19	.26
CuO	.02	.01	.00	.00	—	—	—	—	.05	.05	.00	.00
Na_2O	.07	.04	.03	.06	—	—	—	—	.03	.03	.02	.02
K_2O	.01	.02	.01	.023	—	—	—	—	.02	.02	.01	.014
P_2O_5	—	—	—	—	—	—	—	trace	—	—	—	—
SO_3	.02	.00	.09	.31	—	—	—	.37	.02	.00	.08	.07
CO_2	.07	.14	.01	.02	—	—	—	.10	.00	.11	.04	.06
H_2O-	.83	.65	.28	.60	—	1.64	1.57	—	.60	.26	.24	.32
Total	100.495	100.235	100.113	100.104	99.57	99.99	100.18	98.32	100.61	100.461	100.253	100.701

	P-5	P-6	P-7	P-8	P-9	P-10	P-11	P-12	P-13	P-14	P-15
SiO_2	41.26	40.75	41.30	43.45	43.60	38.40	41.65	44.25	44.50	43.53	44.70
Al_2O_3	2.38	2.51	1.59	.81	1.03	.10	.10	.24	1.41	1.89	.50
Fe_2O_3	2.01	3.47	3.61	.88	.90	3.42	2.88	1.34	none	.49	.07
MgO	38.40	36.94	36.30	41.90	41.00	41.91	41.06	39.02	41.56	37.52	42.05
FeO	3.31	3.12	4.51	.69	.81	none	.16	.60	.35	4.21	.29
MnO	—	—	.11	none	.04	.05	.05	.02	none	.04	—
NiO	—	—	.26	—	.16	—	—	—	none	.20	—
H_2O+	12.70	11.74	11.19	12.29	12.18	15.03	13.10	12.54	12.36	11.69	12.43
TiO_2	—	.06	.15	.02	.01	none	none	.07	none	none	none
Cr_2O_3	—	—	.30	—	.02	—	—	—	.06	.01	—
CaO	—	1.62	.43	.04	.05	none	none	—	.02	none	.12
CuO	—	—	—	—	—	—	—	—	—	—	—
Na_2O	—	—	.04	.05	.01	—	—	—	none	none	—
K_2O	—	—	.02	.02	.03	—	—	—	none	none	—
SO_3	—	—	.06	—	—	—	—	—	—	—	—
CO_2	—	—	.03	—	—	—	—	—	—	—	—
H_2O-	—	.31	—	.04	.08	1.26	1.12	2.06	none	.55	.06
Total	100.06	100.52	99.90	100.19	99.92	100.17	100.12	100.14	100.355	100.13	100.22

TABLE 1. (*Continued*)

	Am	Cr	Fe''-Ch	Fe'''-Ch	Sh	Ri	Ap	Th	Cl	Pe	Da
SiO_2	20.95	16.42	23.81	24.69	27.12	26.50	23.47	24.18	31.18	34.06	22.0
Al_2O_3	35.21	.90	23.12	23.61	27.68	20.85	21.03	18.23	18.28	11.75	27.6
Fe_2O_3	—	29.72	.23	45.61	.20	1.90	1.86	10.56	2.00	1.92	4.7
MgO	22.88	—	2.72	2.74	30.96	19.85	6.72	5.40	31.11	33.90	4.7
FeO	8.28	41.86	39.45	.47	1.24	18.73	35.14	29.75	4.85	2.78	30.2
MnO	trace	—	—	—	.54	.52	.05	.48	.03	—	—
H_2O+	13.02	10.17	10.67	2.88	12.82	11.65	11.23	9.54	12.73	13.08	10.6
TiO_2	—	—	—	—	—	.03	—	—	.10	—	—
Cr_2O_3	—	—	—	—	—	—	—	—	—	4.31	—
CaO	.58	1.32	—	—	—	—	.40	.24	—	—	—
Na_2O	—	—	—	—	—	—	—	—	—	2.45	—
K_2O	—	—	—	—	—	—	—	—	—	.39	—
P_2O_5	—	—	—	—	—	—	—	—	—	—	—
H_2O-	.23	—	—	—	.01	.12	.80	1.07	.10	—	.1
Total	101.15	100.39	100.00	100.00	100.57	100.15	100.70	99.45	100.38	104.64	99.9

	D-1	D-2	D-3	D-4	D-5	D-6	D-7	K-5	K-6	K-7	K-8	K-9
SiO_2	43.10	44.64	45.04	46.35	46.53	46.55	45.4	45.56	44.81	45.44	44.70	44.74
Al_2O_3	40.10	40.42	40.70	39.59	38.93	38.90	39.2	37.65	37.82	38.52	38.64	37.97
Fe_2O_3	.64	.32	trace	.11	—	—	.2	1.35	.92	.80	none	1.44
MnO	none	—	—	—	—	—	none	none	none	—	none	none
MgO	.20	.05	trace	—	—	—	.3	.07	.35	.08	.08	.06
CaO	.24	.34	.22	—	—	—	trace	.10	.43	.08	.24	.09
TiO_2	—	—	trace	—	—	—	none	.19	.37	.16	.22	.27
H_2O-	1.08	.04	none	13.93	14.54	14.04	.4	.76	1.10	.60	.64	.58
H_2O+	14.82	13.98	14.08	—	—	—	13.4	13.66	14.27	13.60	13.88	13.98
CO_2	—	—	—	—	—	—	trace	—	—	—	—	—
P_2O_5	—	—	—	—	—	—	none	—	—	—	—	—
K_2O	—	—	—	—	—	—	.2	.11	—	.14	.14	.16
Na_2O	—	—	—	—	—	—	trace	1.16	—	.66	.62	.76
Total	100.18	99.79	100.04	99.98	100.00	99.49	100.1*	100.61	100.07	100.08	100.12	100.05

* Includes 1.0 carbonaceous matter.

	K-10	K-11	K-12	K-13	K-14	K-15	K-16	H-2	H-3	H-4	H-5	H-6
SiO_2	43.64	44.92	44.06	44.26	43.78	43.1	45.1	44.75	40.26	43.67	44.34	44.08
Al_2O_3	38.33	40.22	39.44	40.22	40.06	34.0	37.7	36.94	37.95	37.91	37.39	39.20
Fe_2O_3	1.43	.54	.80	.30	.64	.67	.7	.31	.30	.26	.42	.10
MnO	—	—	trace	none	—	—	—	—	—	—	—	—
MgO	1.02	.14	.26	.18	.16	—	—	—	—	trace	.04	.05
CaO	1.48	.08	.06	.32	.36	—	—	.11	.22	—	.17	none
K_2O	—	—	—	—	—	—	—	{.60	.74}	—	.04	.20}
Na_2O	—	—	—	—	—	—	—			—	.17	
TiO_2	—	—	—	—	—	5.73	1.4	—	—	—	—	—
H_2O-	.60	.08	1.06	.64	1.02	3.00	.9	2.53	4.45	3.79	2.00	1.44
H_2O+	13.64	14.22	14.16	14.16	14.08	12.30	13.9	14.89	15.94	14.50	15.09	14.74
FeO	—	—	—	—	—	—	—	—	—	—	—	none
P_2O_5	—	—	—	—	—	—	—	—	—	.12	—	—
Total	100.14	100.20	99.84	100.08	100.10	98.80	99.7	100.13	99.86	100.25	99.66	99.81

	H-7	H-8	H-9	H-10	H-11	H-12	H-13	H-21	H-22	H-23	H-24	H-25	H-26
SiO_2	44.08	41.62	44.68	44.50	39.84	44.18	43.10	45.20	44.51	44.35	39.22	43.6	44.3
Al_2O_3	38.60	38.66	38.5	38.68	39.70	39.34	40.10	38.96	39.90	40.35	34.22	40.3	39.1
Fe_2O_3	.32	.62	.39	.24	.64	.32	.64	.21	.21	.21	.10	.4	.4
FeO	—	—	—	—	—	—	—	—	—	—	—	—	—
MgO	.22	.08	.08	.05	.14	.03	.20	.08	.05	.04	.29	—	—
CaO	.12	.10	.18	none	.40	.30	.24	—	—	—	.18	—	—
Na_2O	—	—	.11	1.19	—	—	—	—	—	—	.10	—	—
K_2O	—	—	.05	.14	—	—	—	—	—	—	.09	—	—
P_2O_5	—	—	—	—	—	—	—	—	—	—	—	—	—
H_2O-	2.34	4.26	1.55	1.17	1.76	.96	1.08	—	—	—	13.00	2.5	4.0
H_2O+	14.72	14.64	14.90	14.38	17.52	14.96	14.82	15.35	15.44	15.54	13.00	14.7	13.4
MnO	—	—	—	—	—	—	—	—	—	—	.01	.1	.1
TiO_2	—	—	—	—	—	—	—	—	—	—	<.0001	—	—
Total	100.40	99.98	100.53	100.35	100.00	100.09	100.18	99.80	100.11	100.49	100.21	101.6	101.3

Source Data for Analyses

Chrysotile:

C-1   Fiber, Quebec, Canada. Kalousek and Muttart (1957).
C-2   Fiber, Delaware County, Pa. Kalousek and Muttart (1957).
C-3   Fiber, Aboutville, N. Y. Kalousek and Muttart (1957).
C-4   Fiber, Montville, N. J. Kalousek and Muttart (1957).
C-5   Chrysotile, average of 29 analyses.
C-6   Chrysotile, Gila County, Arizona. J. J. Fahey, analyst. Nagy and Faust (1956).
C-7   Silky Chrysotile, Transvaal. W. A. Deer, analyst. Brindley and Zussman (1957).
C-8   Chrysotile, Woodsreef, Barraba, New South Wales. W. A. Greig, analyst. Proud and Osborne (1952).

Platy serpentine:

P-1(M)   Matrix, Quebec Canada. Kalousek and Muttart (1957).
P-2(M)   Matrix, Delaware County, Pa. Kalousek and Muttart (1957).
P-3(M)   Matrix, Aboutville, N. Y. Kalousek and Muttart (1957).
P-4(M)   Matrix, Montville, N. J. Kalousek and Muttart (1957).
P-5   Antigorite, average of 14 analyses.
P-6   Antigorite, Val Antigorio, Piedmont, Italy. S. Caillere (1936).
P-7   Matrix, Val Antigorio, Italy. Kalousek and Muttart (1957).
P-8   Antigorite, Mikonui, New Zealand. R. A. Howie, analyst. Zussman (1954).
P-9   Antigorite, Caracas, Venezuela. L. C. Peek, analyst. Hess, Smith and Dengo (1952).
P-10   Antigorite No. 2, Nikka Vord Quarries, Shetland Islands. O. v. Knorring, analyst. Brindley and v. Knorring (1954).
P-11   Antigorite No. 1, Nikka Vord Quarries, Shetland Islands. O. v. Knorring, analyst. Brindley and v. Knorring (1954).
P-12   Deweylite, near Murfreesboro, Pike County, Ark. J. J. Fahey, analyst. Nagy and Faust (1956).
P-13   Williamsite, State Line pits, Cecil County, Md. J. J. Fahey, analyst. Nagy and Faust (1956).
P-14   Baltimoreite, Baltimore County, Md. J. J. Fahey, analyst. Nagy and Faust (1956).
P-15   "Yu-Yen Shi Stone," Liaoning Province, Manchuria. J. J. Fahey, analyst. Nagy and Faust (1956).

Other 1:1

Am   Amesite, Chester, Mass. E. V. Shannon (1921)
Cr   Cronstedtite, Kisbanya, Hungary. B. Gossner (1935).
Fe''-Ch   Ferrous chamosite, Northamptonshire, England. R. F. Youell, analyst. Brindley and Youell (1953).
Fe'''-Ch   Ferric chamosite prepared from Fe'''-chamosite by heating in air at 400° C. for two hours. Brindley and Youell (1953).

Chlorites:

Sh   Sheridanite, Savoie, France. Orcel (1937).
Ri   Ripidolite, Androta, Madagascar. Orcel (1927).
Ap   Aphrosiderite, Weilburg, Nassau, Germany. Orcel (1927).
Th   Thuringite, Evisa, Corsica. Orcel (1927).

Cl Clinochlore, Besafotre, Madagascar. Orcel (1927).
Pe Pennine, Zermatt, Switzerland. R. Schlaepfer, analyst. Orcel (1927).
Da Daphnite, Cornwall, England. R. F. Youell, analyst. Brindley and Gillery (1954).

Dickites:

D-1 Dickite, Neurode, Silesia. F. A. Gonyer, analyst. Ross and Kerr (1930).
D-2 Dickite, Greenwood, Ark. F. A. Gonyer, analyst. Ross and Kerr (1930).
D-3 Dickite, Cusihuiriachic, Chihuahua. J. G. Fairchild, analyst. Ross and Kerr (1930).
D-4 Dickite, National Belle mine, Colo. W. F. Hillebrand, analyst. Ross and Kerr (1930).
D-5 Dickite, Island of Anglesey. Ross and Kerr (1930).
D-6 Dickite, Backbone Mountain, Okla. R. K. Bailey, analyst. Ross and Kerr (1930).
D-7 Dickite, Durham, England. C. O. Harvey, analyst. Dunham, Claringbull and Bannister (1948).

Kaolinites:

K-5 Kaolinite, Sand Hill station, Pontiac, S. C. F. A. Gonyer, analyst. Ross and Kerr (1930).
K-6 Kaolinite, Mexia, Tex. F. A. Gonyer, analyst. Ross and Kerr (1930).
K-7 Kaolinite, Roseland, Va. F. A. Gonyer, analyst. Ross and Kerr (1930).
K-8 Kaolinite, Ione, Amador County, Calif. F. A. Gonyer, analyst. Ross and Kerr (1930).
K-9 Kaolinite, Ione, Amador County, Calif. F. A. Gonyer, analyst. Ross and Kerr (1930).
K-10 Kaolinite, Abatik River, Alaska. F. A. Gonyer, analyst. Ross and Kerr (1930).
K-11 Kaolinite, Jerome, Ariz. F. A. Gonyer, analyst. Ross and Kerr (1930).
K-12 Kaolinite, Saline County, Ark. F. A. Gonyer, analyst. Ross and Kerr (1930).
K-13 Kaolinite, Franklin, N. C. F. A. Gonyer, analyst. Ross and Kerr (1930).
K-14 Kaolinite, Saline County, Ark. F. A. Gonyer, analyst. Ross and Kerr (1930).
K-15 Kaolinite, St. Ives, New South Wales. G. T. See, analyst. Loughnan (1957).
K-16 Kaolinite, Huber, Georgia. G. T. See, analyst. Loughnan (1957).

Halloysites:

H-2 Halloysite, Liege, Belgium. L. T. Richardson, analyst. Ross and Kerr (1934).
H-3 Halloysite, Huron County, Ind. L. T. Richardson, analyst. Ross and Kerr (1934).
H-4 Halloysite, Huron County, Ind. L. T. Richardson, analyst. Ross and Kerr (1934).
H-5 Halloysite, Peppers, N. C. E. T. Erickson, analyst. Ross and Kerr (1934).
H-6 Halloysite, Hickory, N. C. J. G. Fairchild, analyst. Ross and Kerr (1934).
H-7 Halloysite, Hickory, N. C. F. A. Gonyer, analyst. Ross and Kerr (1934).
H-8 Halloysite, Adams County, Ohio. F. A. Gonyer, analyst. Ross and Kerr (1934).
H-9 Halloysite, Brandon, Rankin County, Miss. Charles Milton, analyst. Ross and Kerr (1934).

H-10   Halloysite, Leakey, Real County, Tex. R. K. Bailey, analyst. Ross and Kerr (1934).

H-11   Halloysite, Sneeds Creek, Newton County, Ark. F. A. Gonyer, analyst. Ross and Kerr (1934).

H-12   Halloysite, Franklin, N. C. F. A. Gonyer, analyst. Ross and Kerr (1934).

H-13   Halloysite, Myeline, Saxony. F. A. Gonyer, analyst. Ross and Kerr (1934).

H-21   Endellite, Djebal Deber, Libya dried at 110° C. L. T. Alexander, analyst. Alexander et al. (1943).

H-22   Endellite, Anamosa, Iowa dried at 110° C. L. T. Alexander, analyst. Alexander et al. (1943).

H-23   Endellite, Eureka, Utah dried at 110° C. L. T. Alexander, analyst. Alexander et al. (1943).

H-24   Hydrated halloysite, Ness County, Kansas. Swineford et al. (1954).

H-25   Halloysite, Eureka, Utah. G. T. See, analyst. Loughnan (1957).

H-26   Halloysite, Bedford, Indiana. G. T. See, analyst. Loughnan (1957).

fact that misfit of the two sheets that make up the layer can and apparently does occur, the ratio of the number of cation positions in the tetrahedral versus the octahedral sheet may vary from the commonly accepted value of 2:3. Any resulting departure from electrostatic balance may be corrected by appropriate distribution of H^+ in the layer. In addition to this underlying premise the following assumptions are made:

1) of the elements listed in the chemical analyses only Si, Al, Fe, Mg, Mn, Ni, O, and H are considered for positions in the mineral structures discussed here;

2) the total cation charge is adjusted to balance a negative charge resulting from 18 oxygen ions;

3) all silicon is allocated to the tetrahedral sheet (IV) and all divalent cations are placed in the octahedral sheet (VI);

4) except in the case of kaolin minerals, Al and Fe''' are divided equally between IV and VI independent of the amount of Si and Mg present (see Zussman, 1954, p. 510; and Roy, 1952); in the kaolin minerals any Al and Fe''' in excess of the number of Si ions is divided equally between IV and VI. Neglecting small amounts of divalent cations in the kaolin minerals this is the equivalent of assuming that any excess negative charge resulting from substitution for silicon in the tetrahedral sheet will be balanced by an equal positive charge due to substitution in the octahedral sheet.

The resulting procedure yields structural formulas of the usual type in which all the cations except hydrogen are appropriately allocated. Although much of the following discussion pertains only to the distribution of cations other than hydrogen, two additional steps have been added to

the calculations in order to provide information as to a logical distribution of H^+, $(OH)^-$, and O^- in the lattice. 1) Oxygen and hydroxyl ions are proportioned to correspond with the following ideal structural proportions by simply assigning the proper number to account for the sum of the cation charges in each sheet:

$$\frac{|\ O_6Si_4O_2\ |}{IV} \qquad \frac{|\ O_2(OH)_2Mg_6(or\ Al_4)(OH)_6\ |}{VI}$$
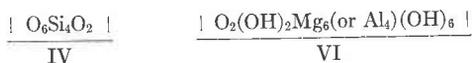
2) Since the amount of O and OH required for this procedure is usually less than and sometimes greater than that available from the analysis, a final adjustment is required involving either a) the subtraction of H^+ and consequent adjustment of associated oxygen in the octahedral sheet or b) the addition of the extra H^+ and associated oxygen. The latter is accomplished by adding the H to the tetrahedral sheet as $(H_4)^{4+}$ simply because, in the opinion of the author, this is the least involved procedure to follow until experimental data reveal the actual proton positions.

The examples in Table II illustrate the complete procedure. Table III gives the number of each of the various cations in a structure unit containing 18 oxygens. Table IV gives the structural assignments prior to readjustments for excess or deficient hydrogen and oxygen. Table V records the complete structural formula after all adjustments have been made.

Relationships of Morphology to Chemistry

General Statement

The factors that determine the degree of curvature and therefore the shape of the minerals considered here are: 1) the misfit of the tetrahedral and octahedral sheets which make up the 1:1 layer; and 2) the strength of the interlayer bonds. These factors, in turn, are affected by: a) the number of cations (and associated oxygens) in the tetrahedral sheet relative to those in the octahedral sheet; b) the size and polarizing power of the cations; and c) the distribution of H^+ ions.

With these factors in mind certain general observations are pertinent. First, interlayer bonds are stronger in the dioctahedral minerals than in the trioctahedral varieties because the Al has a greater effect than Mg on the adjacent OH and therefore on the resulting creation of hydroxyl bonds.

Second, subsitution of Al or Fe''' for Si tends to make the tetrahedral sheet larger, whereas substitution of the same ions for Mg will make the octahedral sheet smaller and at the same time tend to strengthen interlayer bonds in accord with the reasoning in the preceding paragraph. Because cation substitution is more common in the trioctahedral than in

TABLE II. EXAMPLES OF FORMULA CALCULATIONS

Example #1: Kaolinite #10, Abatik River, Alaska.

Analysis:	SiO_2 43.64	Al_2O_3 38.33	Fe_2O_3 1.43	MgO 1.02	H_2O+ 13.64	H_2O- 0.60
Cationic equivalents:	3.874	3.992	.096	.135	8.038	
Cation allocation: IV:	$Si_{3.874}$	$Al_{.059}$				
VI:	$Al_{3.874+.059}$	$Fe_{.048}'''$	$Fe_{.048}'''$	$Mg_{.135}$		
	(charge=15.748)			(charge=12.214)		

Allocation of H^+ and O:

A. Proportioning in accord with cation distribution:

IV: Oxygens in proportion 6:2 to account for 15.748 positive charge: $O_{5.905}:O_{.968}$

VI: Oxygens and (OH) in proportion $O_2:(OH)_2:(OH)_6$ to account for 12.214 positive charge: $O_{2.036}:(OH)_{2.036}:(OH)_{6.107}$

Total oxygen used: 18.052. Resulting oxygen deficiency: $18.052-18.00=.052$ ions

Total hydrogen used: 8.143. Resulting hydrogen deficiency: $8.143-8.038=.105$ ions

B. Adjustment in octahedral sheet to account for deficiencies: Removal of .105 H^+ and .052 $O^=$ from VI gives $O_{2.088}:(OH)_{2.010}:(OH)_{6.028}$

Complete structural formula:

$$O_{5.905}(Si_{3.874}Al_{.059})'''O_{1.968+2.088}(OH)_{2.010}(Al_{3.874+.059}Fe_{.048}'''Mg_{.135})(OH)_{6.028}$$

Example #2: Chrysotile #6, Gila County, Arizona.

Analysis:	SiO_2 42.02	Al_2O_3 0.52	Fe_2O_3 0.19	MgO 41.44	FeO 0.11	MnO 0.03	H_2O+ 14.04	H_2O- 1.64
Cationic equivalents:	3.901	.058	.013	5.733	.008	.003	8.694	
Cation allocation: IV:	$Si_{3.901}$	$Al_{.029}$						
VI:	$Mg_{5.733}$	$Al_{.029}$	$Fe_{.006}'''$	$Fe_{.007}'''$	$Fe_{.008}''$	$Mn_{.003}$		
	(charge=15.711)				(charge=11.595)			

Allocation of H^+ and O:

A. Proportioning in accord with cation distribution:

IV: Oxygens in proportion 6:2 in account for 15.711 positive charge: $O_{5.892}:O_{1.964}$

VI: Oxygens and (OH) in proportion $O_2:(OH)_2:(OH)_6$ to account for 11.595 positive charge: $O_{1.932}:(OH)_{1.932}:(OH)_{5.797}$

Total oxygen used: 17.517. Resulting oxygen excess: $18.000-17.517=.483$ ions

Total hydrogen used: 7.729. Resulting hydrogen excess: $8.694-7.729=.965$ ions

B. Adjustment in tetrahedral sheet to account for excess:

Addition of excess H^+ gives: $Si_{3.901}Al_{.029}Fe_{.007}'''(H_4)_{.241}$

Addition of excess O in proportion 6:2 gives: $O_{6.254}:O_{2.085}$

Complete structural formula:

$$O_{6.254}(Si_{3.901}Al_{.029}Fe_{.007}'''(H_4)_{.241})O_{2.085+1.932}(OH)_{1.932}(Mg_{5.733}Al_{.029}Fe_{.007}'''Fe_{.008}''Mn_{.003})(OH)_{5.79?}$$

TABLE III. CATIONS PER STRUCTURE UNIT OF 18 OXYGENS

	C-1	C-2	C-3	C-4	C-5	C-6	C-7	C-8	P-1(M)	P-2(M)	P-3(M)	P-4(M)
Si	3.840	3.936	3.854	3.883	3.905	3.901	3.859	3.924	3.809	3.891	3.860	3.877
Al	.026	.035	.011	.020	.037	.058	.088	.006	.062	.081	.030	.054
Fe'''	.020	.035	.048	.056	.037	.013	.089	.171	.026	.033	.065	.103
Mg	5.876	5.863	5.881	5.717	5.654	5.733	5.756	5.445	5.849	5.887	5.870	5.778
Fe''	.003	—	.003	.003	.056	.008	.050	.170	.005	.005	.005	.005
Mn	.001	.003	.003	.003	—	.003	.003	.012	.003	.008	.003	.003
Ni	—	—	—	—	—	—	—	—	—	—	—	—
H+	8.739	8.312	8.633	8.794	8.734	8.694	8.501	8.514	8.784	8.290	8.547	8.449

	P-5	P-6	P-7	P-8	P-9	P-10	P-11	P-12	P-13	P-14	P-15
Si	3.855	3.909	4.019	4.047	4.082	3.574	3.914	4.172	4.094	4.131	4.133
Al	.262	.284	.183	.089	.114	.011	.011	.026	.153	.211	.054
Fe'''	.224	.250	.265	.062	.064	.239	.203	.094	.340	.006	.006
Mg	5.355	5.277	5.262	5.811	5.718	5.811	5.743	5.481	5.694	5.303	5.791
Fe''	.252	.250	.368	.053	.064	—	.011	.048	.027	.333	.022
Mn	—	—	.008	—	.003	.003	.003	.003	—	.003	—
Ni	—	—	.021	—	.011	—	—	—	.069	.031	—
H+	7.906	7.708	7.258	7.630	7.545	9.325	8.199	7.883	7.581	7.398	7.663

	Am	Cr	Fe''-Ch	Fe'''-Ch	Sh	Ri	Ap	Th	Cl	Pe	Da
Si	2.000	2.160	2.656	2.836	2.259	2.703	2.590	2.760	2.958	3.269	2.390
Al	3.960	.129	3.036	3.197	2.717	2.505	2.736	2.453	2.043	1.330	3.532
Fe'''	—	2.941	.020	3.942	1.253	.145	.155	.908	.142	.138	.385
Mg	3.254	—	.452	.569	3.843	3.017	1.104	.919	4.394	4.849	.760
Fe''	.662	4.600	3.678	.045	.085	1.595	3.242	2.839	.384	.222	2.744
Mn	—	—	—	—	.037	.046	.003	.048	.003	—	—
Ni	—	—	—	—	—	—	—	—	—	—	—
H+	8.286	8.917	7.933	2.208	7.121	7.920	8.267	7.263	8.049	8.373	7.680

	D-1	D-2	D-3	D-4	D-5	D-6	D-7	K-5	K-6	K-7	K-8	K-9
Si	3.738	3.868	3.880	3.988	3.982	4.015	3.978	4.004	3.923	3.983	3.922	3.930
Al	4.099	4.127	4.132	4.013	3.925	3.955	4.046	3.899	3.902	3.979	3.994	3.930
Fe'''	.042	.031	—	.007	—	—	.014	.090	.061	.053	.063	.095
Mg	.026	.005	—	—	—	—	.039	.008	.044	.010	.010	.008
H+	8.571	8.076	8.086	7.988	8.296	8.077	7.829	8.002	8.329	7.951	8.120	8.188

	K-10	K-11	K-12	K-13	K-14	K-15	K-16	H-2	H-3	H-4	H-5	H-6
Si	3.874	3.866	3.844	3.841	3.781	4.125	3.980	3.933	3.603	3.872	3.881	3.838
Al	3.992	4.078	4.054	4.112	4.077	3.835	3.920	3.826	4.004	3.961	3.857	4.021
Fe'''	.096	.034	.052	.019	.041	.048	.046	.021	.020	.018	.028	.007
Mg	.135	.018	.034	.023	.205	—	—	—	—	—	.005	.005
H+	8.038	8.161	8.238	8.193	8.108	7.849	8.178	8.727	9.516	8.575	8.807	8.554

	H-7	H-8	H-9	H-10	H-11	H-12	H-13	H-21	H-22	H-23	H-24	H-25	H-26
Si	3.849	3.724	3.870	3.895	3.427	3.820	3.738	3.859	3.791	3.763	3.862	3.775	3.931
Al	3.971	4.075	3.938	3.990	4.026	4.008	4.099	3.920	4.003	4.035	3.970	4.112	4.088
Fe'''	.021	.041	.026	.016	.041	.021	.042	.014	.014	.014	.008	.026	.027
Mg	.029	.010	.010	.005	.018	.002	.026	.010	.005	.005	.041	—	—
H+	8.569	8.733	8.605	8.390	10.051	8.627	8.571	8.740	8.770	8.791	8.534	8.486	7.931

the dioctahedral minerals considered here, the effect of cation substitution on morphology is more pronounced in the serpentines than in the kaolinites.

Third, with or without substitution of the type considered above, the amount of misfit is affected by the number of ions in the tetrahedral sheet

TABLE IV. STRUCTURAL ASSIGNMENTS

(Prior to adjustment for excess or deficient O and H)

Sample	Tetrahedral Sheet					Octahedral Sheet									Excess or Deficient (−)	
	O	Si	Al	Fe'''	O	O	(OH)	Al	Fe'''	Mg	Fe''	Mn	Ni	(OH)	O	H
Chrysotile:																
C-1	5.785	3.840	.013	.010	1.928	1.972	1.972	.013	.010	5.876	.003	.001	—	5.915	.428	.852
C-2	5.943	3.936	.009	.010	1.981	1.973	1.973	.010	.009	5.863	—	.003	—	5.931	.211	.420
C-3	5.814	3.854	.006	.024	1.938	1.977	1.977	.005	.024	5.881	.003	.003	—	5.931	.363	.725
C-4	5.868	3.883	.010	.028	1.956	1.926	1.926	.010	.028	5.717	.003	.003	—	5.926	.545	1.089
C-5	5.900	3.905	.018	.019	1.966	1.922	1.922	.019	.018	5.654	.056	—	—	5.766	.526	1.046
C-6	5.892	3.901	.029	.006	1.964	1.932	1.932	.029	.007	5.733	.008	.003	—	5.797	.483	.965
C-7	5.888	3.859	.044	.044	1.963	1.966	1.966	.044	.045	5.756	.005	.003	—	5.898	.319	.637
C-8	5.986	3.924	.003	.085	1.995	1.920	1.920	.003	.085	5.445	.170	.012	—	5.760	.419	.838
Platy Serpentine:																
P-1(M)	5.763	3.809	.031	.013	1.921	1.975	1.975	.031	.013	5.849	.005	.003	—	5.924	.442	.885
P-2(M)	5.901	3.891	.041	.016	1.967	1.996	1.996	.040	.017	5.887	.005	.008	—	5.986	.154	.308
P-3(M)	5.838	3.860	.015	.032	1.946	1.981	1.981	.015	.033	5.870	.005	.003	—	5.942	.312	.624
P-4(M)	5.904	3.877	.015	.028	1.968	1.968	1.968	.014	.028	5.778	.005	.003	—	5.904	.288	.577
P-5	6.056	3.855	.131	.112	2.019	1.990	1.990	.131	.112	5.355	.252	—	—	5.971	−.026	−.055
P-6	6.164	3.909	.142	.125	2.054	1.976	1.976	.142	.125	5.277	.368	—	—	5.928	−.098	−.196
P-7	6.282	4.019	.091	.133	2.094	1.998	1.998	.092	.132	5.262	.250	.008	—	5.995	−.367	−.735
P-8	6.156	4.047	.045	.031	2.052	1.993	1.993	.044	.031	5.811	.053	—	.021	5.978	−.172	−.341
P-9	6.222	4.082	.057	.032	2.074	1.977	1.977	.057	.032	5.718	.064	.003	—	5.930	−.180	−.362
P-10	6.502	3.574	.005	.119	1.834	2.001	2.001	.006	.120	5.718	—	.003	.011	6.002	.660	1.322
P-11	5.986	3.914	.006	.101	1.995	1.973	1.973	.005	.102	5.743	.011	.003	—	5.919	.154	.307
P-12	6.327	4.172	.013	.047	2.109	1.874	1.874	.013	.047	5.481	.048	.003	—	5.623	.193	.386
P-13	6.228	4.094	.076	—	2.076	1.969	1.969	.077	—	5.694	.027	—	.069	5.906	−.148	−.294
P-14	6.334	4.131	.105	.170	2.111	1.952	1.952	.106	.170	5.303	.333	.003	.031	5.855	−.204	−.409
P-15	6.232	4.133	.027	.003	2.077	1.953	1.953	.027	.003	5.791	.022	—	—	5.858	−.073	−.148
Other 1:1																
Am	5.227	2.000	1.980	—	1.742	2.295	2.295	1.980	—	3.254	.662	—	—	6.886	−.445	−.895
Cr	4.974	2.160	.070	1.470	1.658	2.303	2.303	.070	1.471	—	4.600	—	—	6.910	−.148	−.296
Fe''-Ch	5.706	2.656	.518	.010	1.902	2.142	2.142	1.518	.010	.452	3.678	—	—	6.425	−.317	−.634
Fe'''-Ch	6.052	2.836	1.599	—	2.017	2.942	2.942	1.598	3.942	.569	.045	—	—	8.825	−4.778	−9.559
Chlorite:																
Sh	5.622	2.259	1.359	.626	1.874	2.314	2.314	1.358	.627	3.843	.085	.037	—	6.943	−1.067	−2.136
Ri	5.544	2.703	1.253	.072	1.848	2.215	2.215	1.252	.073	3.017	1.595	.046	—	6.646	−.468	−.941
Ap	5.511	2.590	1.368	.077	1.837	2.172	2.172	1.368	.078	1.104	3.242	.003	—	6.517	−.209	−.422
Th	6.030	2.760	1.227	.454	2.010	2.109	2.109	1.226	.454	.919	2.839	.048	—	6.327	−.585	−1.173
Cl	5.667	2.958	1.022	.071	1.889	2.140	2.140	1.021	.071	4.394	.384	.003	—	6.421	−.257	−.512
Pe	5.730	3.269	.665	.069	1.910	2.058	2.058	.665	.069	4.849	.222	—	—	6.173	.071	.142
Da	5.788	2.390	1.766	.192	1.929	2.147	2.147	1.766	.193	.760	2.744	—	—	6.442	−.453	−.909

TABLE IV (Continued)

Sample	Tetrahedral Sheet					Octahedral Sheet						Excess or Deficient (−)	
	O	Si	Al	Fe'''	O	O	(OH)	Al	Fe'''	Mg	(OH)	O	H
Dickite:													
D-1	5.835	3.738	.181	.021	1.945	1.978	1.978	3.738+.180	.021	.026	5.934	.330	.659
D-2	5.958	3.868	.129	.010	1.986	2.006	2.006	3.868+.130	.011	.005	6.017	.027	.053
D-3	5.961	3.880	.126	.003	1.987	2.003	2.003	3.880+.126	.004		6.009	.037	.074
D-4	6.000	3.988	.013	—	2.000	2.002	2.002	3.988+.012	—		6.005	−.009	−.019
D-5	5.973	3.982	—	—	1.991	1.963	1.963	3.925	—		5.888	−.222	−.445
D-6	6.022	4.015	—	—	2.007	1.977	1.977	3.955	—	.039	5.932	.085	.168
D-7	6.012	3.978	.034	.007	2.004	2.023	2.023	3.978+.034	.007		6.068	−.130	−.262
Kaolinite:													
K-5	6.006	4.004	—	—	2.002	1.997	1.997	3.899	.090	.008	5.991	.007	.014
K-6	5.907	3.923	—	.020	1.969	1.986	1.986	3.902	.021+.020	.044	5.959	.193	.384
K-7	6.003	3.983	—	.024	2.001	2.007	2.007	3.979	.004+.025	.010	6.022	−.040	−.078
K-8	5.958	3.922	.036	.031	1.983	1.998	1.998	3.922+.036	.032	.010	5.995	.065	.127
K-9	5.949	3.930	—	.047	1.968	1.992	1.992	3.930	.048	.135	6.107	.109	.221
K-10	5.905	3.874	.059	.048	1.979	2.036	2.036	3.866+.059	.017	.034	6.002	−.052	−.105
K-11	5.937	3.866	.106	.017	1.971	2.001	2.001	3.844+.106	.026	.018	5.997	−.080	−.158
K-12	5.913	3.844	.105	.026	1.975	1.999	1.999	3.841+.105	.009	.034	6.003	.121	.242
K-13	5.925	3.841	.135	.010	1.971	2.001	2.001	3.781+.136	.021	.023	6.129	.095	.189
K-14	5.862	3.781	.148	.020	1.954	2.043	2.043	3.781+.148	.048	.205	5.825	−.031	−.064
K-15	6.189	4.125	—	—	2.063	1.942	1.942	3.835	.046	—	5.949	−.039	−.082
K-16	5.970	3.980	—	—	1.990	1.983	1.983	3.920		—		−.125	−.246
Halloysite:													
H-2	5.899	3.933		—	1.966	1.923	1.923	3.826	.021	—	5.770	.519	1.034
H-3	5.643	3.603	.201	.010	1.881	1.906	1.906	3.603+.200	.010	—	5.719	.945	1.891
H-4	5.868	3.872	.045	.009	1.956	1.962	1.962	3.872+.044	.009	.005	5.887	.365	.726
H-5	5.824	3.881		.002	1.941	1.943	1.943	3.857	.024+.002	.005	5.830	.519	1.034
H-6	5.865	3.838	.092	.003	1.955	1.968	1.968	3.838+.091	.004	.029	5.904	.340	.682
H-7	5.853	3.849	.061	.010	1.951	1.970	1.970	3.849+.061	.011	.010	5.910	.346	.689
H-8	5.8-9	3.724	.175	.021	1.935	1.964	1.964	3.724+.176	.020	.010	5.891	.441	.878
H-9	5.907	3.870	.034	.013	1.953	1.962	1.962	3.870+.034	.013	.005	5.886	.378	.757
H-10	5.502	3.895	.048	.008	1.969	1.977	1.977	3.895+.047	.008	.018	5.930	.240	.483
H-11	5.847	3.427	.299	.021	1.834	1.880	1.880	3.427+.300	.020	.002	5.639	1.265	2.532
H-12	5.835	3.820	.094	.010	1.949	1.963	1.963	3.820+.094	.011	.026	5.889	.389	.675
H-13		3.738	.181	.021	1.945	1.978	1.978	3.738+.180	.021		5.934	.330	.659
H-21	5.832	3.859	.031	.007	1.944	1.951	1.951	3.859+.030	.007	.010	5.854	.468	.935
H-22	5.814	3.791	.106	.007	1.938	1.954	1.954	3.791+.106	.007	.005	5.861	.479	.955
H-23	5.805	3.763	.136	.007	1.935	1.955	1.955	3.763+.136	.007	.005	5.864	.486	.972
H-24	5.859	3.862	.054	.004	1.953	1.974	1.974	3.862+.054	.004	.041	5.921	.319	.639
H-25	5.868	3.775	.169	.013	1.956	1.978	1.978	3.775+.168	.013	—	5.934	.286	.574
H-26	6.000	3.931	.079	.013	2.000	2.011	2.011	3.931+.078	.014	—	6.034	−.056	−.114

TABLE V. STRUCTURAL FORMULAS

Chrysotile:

C-1	$O_{6.106}Si_{3.840}Al_{.013}Fe_{.010}'''(H_4)_{.213}O_{2.065}$	$O_{.192}(OH)_{1.973}Mg_{5.876}Al_{.013}Fe_{.010}'''Mn_{.001}Fe_{.003}''(OH)_{5.915}$
C-2	$O_{6.101}Si_{3.836}Al_{.009}Fe_{.010}'''(H_4)_{.105}O_{2.034}$	$O_{.573}(OH)_{1.973}Mg_{5.865}Al_{.010}Fe_{.009}'''Mn_{.003}(OH)_{5.919}$
C-3	$O_{6.086}Si_{3.854}Al_{.006}Fe_{.024}'''(H_4)_{.131}O_{2.059}$	$O_{.977}(OH)_{1.977}Mg_{5.881}Al_{.005}Fe_{.024}'''Mn_{.003}Fe_{.001}''(OH)_{5.931}$
C-4	$O_{6.327}Si_{3.881}Al_{.006}Fe_{.028}'''(H_4)_{.272}O_{2.095}$	$O_{1.976}(OH)_{1.926}Mg_{5.711}Al_{.010}Fe_{.028}'''Mn_{.003}Fe_{.170}''(OH)_{5.981}$
C-5	$O_{6.256}Si_{3.906}Al_{.018}Fe_{.019}'''(H_4)_{.261}O_{2.097}$	$O_{1.925}(OH)_{1.922}Mg_{5.854}Al_{.019}Fe_{.018}'''Fe_{.066}''(OH)_{5.766}$
C-6	$O_{6.264}Si_{3.901}Al_{.029}Fe_{.006}'''(H_4)_{.291}O_{2.085}$	$O_{1.932}(OH)_{1.932}Mg_{5.733}Al_{.029}Fe_{.007}'''Mn_{.003}Fe_{.008}''(OH)_{5.797}$
C-7	$O_{6.127}Si_{3.859}Al_{.044}Fe_{.044}'''(H_4)_{.159}O_{2.043}$	$O_{.960}(OH)_{1.966}Mg_{5.766}Al_{.044}Fe_{.045}'''Mn_{.005}Fe_{.005}''(OH)_{5.598}$
C-8	$O_{6.306}Si_{3.924}Al_{.005}Fe_{.054}'''(H_4)_{.299}O_{2.100}$	$O_{.920}(OH)_{1.950}Mg_{5.445}Al_{.003}Fe_{.035}'''Mn_{.013}Fe_{.170}''(OH)_{5.760}$

Platy Serpentine:

P-1(M)	$O_{6.095}Si_{3.809}Al_{.031}Fe_{.012}'''(H_4)_{.231}O_{2.031}$	$O_{.975}(OH)_{1.973}Mg_{5.849}Al_{.031}Fe_{.013}'''Mn_{.003}Fe_{.005}''(OH)_{5.924}$
P-2(M)	$O_{6.017}Si_{3.891}Al_{.041}Fe_{.016}'''(H_4)_{.077}O_{2.005}$	$O_{.996}(OH)_{1.998}Mg_{5.887}Al_{.040}Fe_{.017}'''Mn_{.008}Fe_{.005}''(OH)_{5.986}$
P-3(M)	$O_{6.072}Si_{3.860}Al_{.015}Fe_{.032}'''(H_4)_{.156}O_{2.024}$	$O_{.961}(OH)_{1.961}Mg_{5.870}Al_{.013}Fe_{.033}'''Mn_{.033}Fe_{.005}''(OH)_{5.942}$
P-4(M)	$O_{6.120}Si_{3.877}Al_{.015}Fe_{.025}'''(H_4)_{.144}O_{2.040}$	$O_{.968}(OH)_{1.968}Mg_{5.778}Al_{.014}Fe_{.055}'''Mn_{.003}Fe_{.005}''(OH)_{5.904}$
P-5	$O_{6.656}Si_{3.855}Al_{.131}Fe_{.112}'''O_{2.019}$	$O_{2.045}(OH)_{1.976}Mg_{5.355}Al_{.131}Fe_{.112}'''Fe_{.252}''(OH)_{5.930}$
P-6	$O_{6.164}Si_{3.909}Al_{.142}Fe_{.125}'''O_{2.054}$	$O_{2.074}(OH)_{1.997}Mg_{6.277}Al_{.142}Fe_{.125}'''Fe_{.250}''(OH)_{5.781}$
P-7	$O_{6.282}Si_{4.019}Al_{.091}Fe_{.133}'''O_{2.094}$	$O_{2.365}(OH)_{1.814}Mg_{5.963}Al_{.092}Fe_{.232}'''Mn_{.008}Fe_{.368}''Ni_{.021}(OH)_{5.444}$
P-8	$O_{6.156}Si_{4.047}Al_{.046}Fe_{.031}'''O_{2.052}$	$O_{.224}(OH)_{1.908}Mg_{6.811}Al_{.044}Fe_{.031}'''Fe_{.053}''(OH)_{5.722}$
P-9	$O_{6.252}Si_{4.082}Al_{.057}Fe_{.032}'''O_{2.074}$	$O_{.157}(OH)_{1.887}Mg_{5.718}Al_{.057}Fe_{.052}'''Mn_{.002}Fe_{.064}''Ni_{.011}(OH)_{6.658}$
P-10	$O_{5.997}Si_{3.674}Al_{.006}Fe_{.119}'''(H_4)_{.300}O_{1.999}$	$O_{2.001}(OH)_{2.001}Mg_{5.811}Al_{.006}Fe_{.129}'''Mn_{.003}(OH)_{6.002}$
P-11	$O_{6.105}Si_{3.914}Al_{.006}Fe_{.101}'''(H_4)_{.077}O_{2.035}$	$O_{.975}(OH)_{1.973}Mg_{5.742}Al_{.005}Fe_{.102}'''Mn_{.003}Fe_{.011}''(OH)_{5.519}$
P-12	$O_{6.472}Si_{4.172}Al_{.013}Fe_{.047}'''(H_4)_{.096}O_{2.757}$	$O_{1.874}(OH)_{1.874}Mg_{5.681}Al_{.013}Fe_{.047}'''Mn_{.003}Fe_{.048}''(OH)_{5.625}$
P-13	$O_{6.228}Si_{4.094}Al_{.076}Fe_{.076}''O_{2.070}$	$O_{2.117}(OH)_{1.846}Mg_{5.694}Al_{.076}Fe_{.027}''Ni_{.069}(OH)_{5.685}$
P-14	$O_{6.334}Si_{4.131}Al_{.106}Fe_{.170}'''O_{2.111}$	$O_{2.156}(OH)_{1.350}Mg_{5.105}Al_{.106}Fe_{.170}'''Mn_{.003}Fe_{.331}''Ni_{.031}(OH)_{5.548}$
P-15	$O_{6.232}Si_{4.133}Al_{.027}Fe_{.003}''O_{2.077}$	$O_{5.026}(OH)_{1.916}Mg_{5.791}Al_{.022}Fe_{.003}''Fe_{.022}''(OH)_{5.747}$

Other 1:1

Am	$O_{5.527}Si_{2.000}Al_{1.986}O_{1.742}$	$O_{2.740}(OH)_{2.071}Al_{1.071}Al_{.980}Mg_{3.254}Fe_{.662}''(OH)_{6.215}$

Table V (*continued*)

Cr	$O_{4.974}Si_{2.160}Al_{.070}Fe_{1.470}'''O_{1.458}$	$O_{1.896}(OH)_{2.222}Al_{.070}Fe_{1.471}'''Fe_{4.600}''(OH)_{6.488}.$
Fe''-Ch	$O_{5.706}Si_{2.656}Al_{.438}Fe_{.010}'''O_{1.902}$	$O_{2.459}(OH)_{1.284}Al_{.518}Fe_{.010}'''Mg_{.462}Fe_{3.678}''(OH)_{5.940}.$
Fe'''-Ch	$O_{6.052}Si_{2.536}Al_{.599}O_{2.017}$	$O_{1.699}(OH)_{.532}Al_{.598}Fe_{3.342}'''Mg_{.560}Fe_{.043}''(OH)_{.650}O_{1.096}$
Chlorites:		
Sh	$O_{5.622}Si_{2.239}Al_{.319}Fe_{.626}'''O_{1.374}$	$O_{2.381}(OH)_{1.780}Mg_{2.843}Al_{.333}Fe_{.027}'''Mn_{.037}Fe_{.085}''(OH)_{5.341}$
Ri	$O_{5.544}Si_{2.703}Al_{.243}Fe_{.072}'''O_{1.546}$	$O_{2.683}(OH)_{1.930}Mg_{3.017}Al_{.252}Fe_{.073}'''Fe_{1.595}''Mn_{.046}(OH)_{5.940}.$
Ap	$O_{5.511}Si_{2.590}Al_{.388}Fe_{.077}'''O_{1.537}$	$O_{2.381}(OH)_{2.667}Mg_{1.104}Al_{.368}Fe_{.078}'''Fe_{3.242}''Mn_{.003}(OH)_{6.200}.$
Th	$O_{6.030}Si_{2.760}Al_{.227}Fe_{.454}'''O_{2.010}$	$O_{2.694}(OH)_{1.316}Fe_{1.839}''Al_{.225}Fe_{.454}''Mg_{.919}Mn_{.058}(OH)_{5.447}$
Cl	$O_{5.667}Si_{2.958}Al_{.025}Fe_{.071}'''O_{1.889}$	$O_{2.397}(OH)_{2.012}Mg_{3.394}Al_{.021}Fe_{.071}''Fe_{.384}''Mn_{.063}(OH)_{6.037}$
Pe	$O_{5.783}Si_{2.989}Al_{.663}Fe_{.069}'''(H_4)_{.035}O_{1.978}$	$O_{2.058}(OH)_{2.058}Mg_{.849}Al_{.605}Fe_{.069}'''Fe_{.222}''(OH)_{6.173}$
Da	$O_{5.788}Si_{2.399}Al_{.766}Fe_{.192}'''O_{1.929}$	$O_{2.600}(OH)_{1.990}Fe_{2.744}''Al_{.796}Fe_{.193}'''Mg_{.780}(OH)_{5.760}$
Dickite:		
D-1	$O_{6.063}Si_{3.738}Al_{.181}Fe_{.021}'''(H_4)_{.165}O_{2.057}$	$O_{1.978}(OH)_{1.978}Al_{3.738+.180}Fe_{.021}'''Mg_{.028}(OH)_{5.934}$
D-2	$O_{5.978}Si_{3.498}Al_{.125}Fe_{.010}'''(H_4)_{.013}O_{1.993}$	$O_{2.006}(OH)_{2.006}Al_{3.368+.120}Fe_{.011}'''Mg_{.005}(OH)_{6.017}$
D-3	$O_{5.989}Si_{3.889}Al_{.126}(H_4)_{.013}O_{1.996}$	$O_{2.003}(OH)_{2.003}Al_{3.880+.158}(OH)_{6.009}$
D-4	$O_{6.000}Si_{3.988}Al_{.013}Fe_{.003}'''O_{2.000}$	$O_{2.011}(OH)_{1.997}Al_{3.988+.012}Fe_{.004}'''(OH)_{5.991}$
D-5	$O_{6.140}Si_{3.982}(H_4)_{.111}O_{2.046}$	$O_{1.962}(OH)_{1.965}Al_{3.935}(OH)_{3.388}$
D-6	$O_{6.066}Si_{4.015}(H_4)_{.092}O_{2.028}$	$O_{1.977}(OH)_{1.977}Al_{3.955}(OH)_{5.953}$
D-7	$O_{6.013}Si_{3.975}Al_{.034}Fe_{.007}'''O_{.004}$	$O_{2.107}(OH)_{1.958}Al_{3.975+.034}Fe_{.007}'''Mg_{.039}(OH)_{5.871}$
Kaolinite:		
K-5	$O_{6.011}Si_{4.004}(H_4)_{.005}O_{2.004}$	$O_{1.997}(OH)_{1.997}Al_{3.898}Fe_{.090}'''Mg_{.038}(OH)_{5.991}$
K-6	$O_{6.052}Si_{3.983}Fe_{.020}'''(H_4)_{.096}O_{2.017}$	$O_{1.986}(OH)_{1.986}Al_{3.902}Fe_{.021+.020}'''Mg_{.044}(OH)_{5.959}$
K-7	$O_{6.003}Si_{3.983}Fe_{.024}'''O_{2.001}$	$O_{2.041}(OH)_{1.988}Al_{3.979}Fe_{.004+.025}'''Mg_{.010}(OH)_{5.965}$
K-8	$O_{6.017}Si_{3.923}Al_{.046}Fe_{.041}'''(H_4)_{.052}O_{2.002}$	$O_{1.998}(OH)_{1.998}Al_{3.922+.046}Fe_{.032}'''Mg_{.010}(OH)_{5.995}$
K-9	$O_{6.031}Si_{3.989}Fe_{.047}'''(H_4)_{.043}O_{2.010}$	$O_{1.992}(OH)_{1.992}Al_{3.920}Fe_{.045}'''Mg_{.008}(OH)_{5.975}$
K-10	$O_{5.965}Si_{3.971}Al_{.059}Fe_{.043}'''O_{1.988}$	$O_{2.038}(OH)_{2.010}Al_{3.874+.059}Fe_{.048}'''Mg_{.135}(OH)_{6.028}$

Table V (Continued)

K-11	$O_{6.097}Si_{3.866}Al_{.106}Fe_{.017}'''(H_4)_{.039}O_{1.999}$	$O_{2.001}(OH)_{2.001}Al_{3.866+.106}Fe_{.017}'''Mg_{.013}(OH)_{6.002}$
K-12	$O_{6.004}Si_{3.844}Al_{.105}Fe_{.026}'''(H_4)_{.060}O_{2.001}$	$O_{1.999}(OH)_{1.999}Al_{3.844+.105}Fe_{.026}'''Mg_{.034}(OH)_{5.997}$
K-13	$O_{5.996}Si_{3.841}Al_{.135}Fe_{.010}'''(H_4)_{.047}O_{1.999}$	$O_{2.001}(OH)_{2.001}Al_{3.841+.135}Fe_{.009}'''Mg_{.023}(OH)_{6.003}$
K-14	$O_{5.862}Si_{3.781}Al_{.148}Fe_{.020}'''O_{1.954}$	$O_{2.074}(OH)_{2.027}Al_{3.781+.148}Fe_{.021}'''Mg_{.205}(OH)_{6.081}$
K-15	$O_{6.219}Si_{4.125}(H_4)_{.090}O_{2.073}$	$O_{1.942}(OH)_{1.942}Al_{3.805}Fe_{.038}'''(OH)_{5.825}$
K-16	$O_{6.064}Si_{3.980}(H_4)_{.061}O_{2.021}$	$O_{1.983}(OH)_{1.983}Al_{3.920}Fe_{.846}'''(OH)_{5.949}$

Halloysite:

H-2	$O_{6.288}Si_{3.933}(H_4)_{.258}O_{2.396}$	$O_{1.923}(OH)_{1.923}Al_{3.856}Fe_{.021}'''(OH)_{5.470}$
H-3	$O_{6.252}Si_{3.603}Al_{.201}Fe_{.010}'''(H_4)_{.473}O_{2.115}$	$O_{1.906}(OH)_{1.906}Al_{3.603+.200}Fe_{.010}'''(OH)_{5.719}$
H-4	$O_{6.142}Si_{3.512}Al_{.045}Fe_{.069}'''(H_4)_{.181}O_{2.047}$	$O_{1.962}(OH)_{1.962}Al_{3.872+.044}Fe_{.009}'''(OH)_{5.887}$
H-5	$O_{6.213}Si_{3.881}Fe_{.002}'''(H_4)_{.268}O_{.072}$	$O_{1.943}(OH)_{1.943}Al_{3.857}Fe_{.024+.002}'''Mg_{.005}(OH)_{5.83C}$
H-6	$O_{6.129}Si_{3.838}Al_{.092}Fe_{.003}'''(H_4)_{.176}O_{2.046}$	$O_{1.908}(OH)_{1.908}Al_{3.838+.091}Fe_{.064}'''Mg_{.005}(OH)_{5.904}$
H-7	$O_{6.113}Si_{3.849}Al_{.061}Fe_{.010}'''(H_4)_{.172}O_{2.051}$	$O_{1.970}(OH)_{1.970}Al_{3.849+.061}Fe_{.011}'''Mg_{.029}(OH)_{5.91v.}$
H-8	$O_{6.136}Si_{3.724}Al_{.175}Fe_{.051}'''(H_4)_{.219}O_{2.045}$	$O_{1.964}(OH)_{1.964}Al_{3.724+.176}Fe_{.020}'''Mg_{.010}(OH)_{5.891}$
H-9	$O_{6.139}Si_{3.870}Al_{.034}Fe_{.013}'''(H_4)_{.189}O_{2.047}$	$O_{1.962}(OH)_{1.962}Al_{3.870+.034}Fe_{.013}'''Mg_{.010}(OH)_{5.886}$
H-10	$O_{5.967}Si_{3.895}Al_{.048}Fe_{.008}'''(H_4)_{.121}O_{2.009}$	$O_{1.977}(OH)_{1.977}Al_{3.895+.047}Fe_{.008}'''Mg_{.005}(OH)_{5.900}$
H-11	$O_{6.451}Si_{3.827}Al_{.599}Fe_{.021}'''(H_4)_{.633}O_{2.156}$	$O_{1.880}(OH)_{1.886}Al_{3.827+.209}Fe_{.029}'''Mg_{.018}(OH)_{5.629}$
H-12	$O_{6.139}Si_{3.826}Al_{.094}Fe_{.010}'''(H_4)_{.169}O_{2.046}$	$O_{1.963}(OH)_{1.963}Al_{3.820+.094}Fe_{.011}'''Mg_{.002}(OH)_{5.889}$
H-13	$O_{6.068}Si_{3.738}Al_{.181}Fe_{.021}'''(H_4)_{.165}O_{2.057}$	$O_{1.978}(OH)_{1.978}Al_{3.738+.180}Fe_{.021}'''Mg_{.026}(OH)_{5.934}$
H-21	$O_{6.183}Si_{3.850}Al_{.031}Fe_{.007}'''(H_4)_{.234}O_{2.083}$	$O_{1.951}(OH)_{1.951}Al_{3.859+.030}Fe_{.007}'''Mg_{.010}(OH)_{5.864}$
H-22	$O_{6.173}Si_{3.791}Al_{.106}Fe_{.007}'''(H_4)_{.239}O_{2.056}$	$O_{1.954}(OH)_{1.954}Al_{3.791+.106}Fe_{.007}'''Mg_{.00C}(OH)_{5.861}$
H-23	$O_{6.170}Si_{3.763}Al_{.136}Fe_{.097}'''(H_4)_{.243}O_{2.056}$	$O_{1.955}(OH)_{1.955}Al_{3.763+.136}Fe_{.007}'''Mg_{.006}(OH)_{5.864}$
H-24	$O_{6.098}Si_{3.862}Al_{.054}Fe_{.004}'''(H_4)_{.160}O_{2.033}$	$O_{1.974}(OH)_{1.974}Al_{3.862+.054}Fe_{.004}'''Mg_{.041}(OH)_{5.921}$
H-25	$O_{6.064}Si_{3.775}Al_{.169}Fe_{.013}'''(H_4)_{.145}O_{2.027}$	$O_{1.978}(OH)_{1.982}Al_{3.931+.6/3}Fe_{.014}'''(OH)_{5.084}$
H-26	$O_{6.000}Si_{3.931}Al_{.079}Fe_{.013}'''O_{5.000}$	$O_{2.067}(OH)_{1.982}Al_{3.931+.6/3}Fe_{.014}'''(OH)_{5.948}$

relative to a given number in the octahedral sheet. Thus, a deficiency or excess of silicon relative to aluminum or magnesium (electrostatic balance being achieved by appropriate H^+ distribution) may play a very important role in affecting morphology.

Fourth, the amount and distribution of H^+ may be very important in affecting the strength of interlayer bonds.

The fact that the greatest chemical variation accompanying the morphological change from kaolinite to halloysite ($4H_2O$) to allophane is that of H^+ content suggests that factor four plays the predominant role in determining the morphology of kaolin-group minerals. On the other hand, factors two and three as well as four are important in the case of the serpentines. Obviously, interaction of all four factors is involved in all cases.

Unfortunately, data are not sufficient to permit a detailed evaluation of these factors and all possible interactions. For this discussion the chemical-morphological relationships have been evaluated in two ways which are believed to relate most directly to the concepts expressed above and which involve the minimum number of assumptions. These two evaluations are demonstrated 1) by plotting the position of the various platy and fibrous minerals with respect to the average size of the cations in the tetrahedral sheet as compared to the average size of those in the octahedral sheet; and 2) by plotting the position of the kaolin group minerals on a SiO_2-Al_2O_3-H_2O diagram and the serpentine minerals on a SiO_2-$R''O$-H_2O diagram (where $R'' = Mg$, Fe'', Mn, and Ni).

Morphology in Relation to Average Radii of Octahedral and Tetrahedral Cations

Figure 2 illustrates the relationship of morphology to the size of the cations in the octahedral versus the tetrahedral sheets of the 1:1 layer structure. The points for chrysotile (C) and kaolinite (K) represent the ideal serpentine and kaolinite compositions whereas those for platy serpentine (PS) and halloysite (H) represent average values obtained from all the analyses of these minerals. Variation within the two groups will be discussed later. The radii used are those of Goldschmidt. The symbols denote whether the compound has tubular or platy crystals.

The position of each compound is determined from the structural assignments in Table IV by plotting the average size of the cations in the tetrahedral sheet against the average size of those in the octahedral sheet. It is, of course, appreciated that "size" does not pertain to misfit of the sheets in the sense of "space occupied by the cation" but is used as a convenient index of the effect of cations of differing polarizing power on the anions which make up the lattice framework.
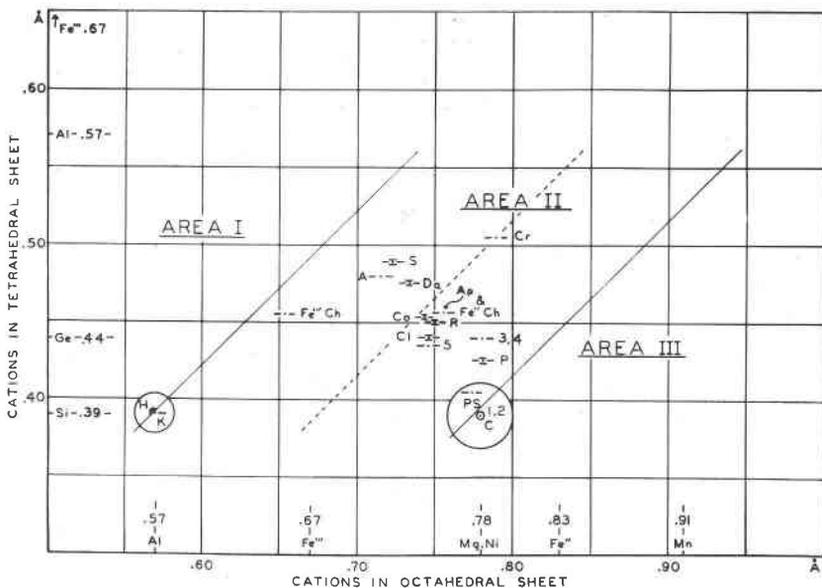
Fig. 2. 1:1 minerals plotted on the basis of the average radius of tetrahedral versus average radius of octahedral cations. (Radii of Goldschmidt.)

A: amesite; Co: corundophyllite (characteristic composition); P: penninite (characteristic composition); PS: platy serpentine; S: sheridanite; Ri: ripidolite. Other initials are the same as those given in the tables.

The figure can be divided into three areas on the basis of the morphology of the compounds. Since there are not enough compounds to determine the precise boundaries, the light lines are arbitrarily drawn from points adjacent to the positions of Si vs. Al and Si vs. Mg at an angle of 45° to the horizontal. It is important to note that the utility of the graph is not dependent upon the validity of the assumptions as to the distribution of cations in the two sheets of the layer. For example, the addition of Al or Fe''' to the ideal serpentine composition will place the position of the resulting compound in area II independent of whether these ions substitute in the tetrahedral sheet, the octahedral sheet, or both.

Area II contains those compounds that occur as platy to flaky crystals. This characteristic morphology is attributed to the fact that in most of this area the size, charge, and number of the ions in octahedral as opposed to tetrahedral coordination is such that misfit between the two sheets of the 1:1 layer is at a minimum and there is little tendency for curvature. On approaching the edges of this region, interlayer bond strength, which is not taken direct account of in this graph, plays an increasingly dominant role.

In the region designated as area I, the chemical composition is such that the "gibbsite" sheet becomes too small for the silicon-oxygen sheet and therefore when interlayer bonds are sufficiently weak tubes having a "negative"* curvature result (silicon-oxygen sheet outside, gibbsite sheet inside). Halloysite ($4H_2O$) is the only natural tubular mineral known from this area and thus far efforts to synthesize this and other tubes in this composition region have been unsuccessful. Indeed it is unlikely that other 1:1 minerals exist in this region because 1) aluminum represents the smallest ion commonly found in the octahedral sheet, and 2) a dioctahedral mineral having appreciable aluminum or ferric iron substituting for silicon in the tetrahedral sheet would be likely to have a 2:1 structure with interlayer cations to compensate for the excess negative charge.

In area III, as a result of the increase of size of "octahedral" as opposed to "tetrahedral" ions, the "brucite" sheet is too large for the silicon-oxygen sheet and tubes having a "positive" curvature result. Natural and synthetic chrysotile and nickel chrysotile ($Si_4Ni_6O_{10}(OH)_8$) represent the only compounds definitely known to occur in this area. (Sufficiently detailed chemical and morphological data on greenalite ($Si_4Fe'''Fe_{4.5}O_{10}(OH)_8$) have not been obtained.) Replacement of part of the Mg or Ni by Fe'' or Mn would result in a still greater misfit of tetrahedral and octahedral sheets and therefore finer tubes of smaller radii of curvature. On the basis of similar reasoning, it is unlikely that many other 1:1 layer silicates occur in area III for, as the radius of curvature is decreased, physical and chemical instability of the extremely thin tubes would result. Previous studies (Nagy and Bates, 1952) have demonstrated that chrysotile itself is very much more susceptible to chemical attack than platy serpentine.

On the basis of present knowledge the two circled areas on the graph encompassing the kaolin and serpentine groups are the only locations where both tubes and plates occur. It seems evident that in both situations naturally-occurring lath-shaped crystals with varying "tendencies to curve" and of varying length/width ratio occupy the area between platy and tubular morphological "end-members." It has already been shown in the case of serpentine (Zussman, 1954; Whittaker and Zussman, 1956; Zussman, Brindley and Comer, 1957; Zussman and Brindley, 1957), and to a lesser extent in the kaolinites (Honjo, Kitamura and Mihama, 1954), that various "structure states" may exist in this transition region.

In order to provide a measure of the relative position of the various

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* The terms positive and negative have no significance except to designate opposite directions of curvature.

compounds on the graph, Table VI lists the calculated perpendicular distance of each from the dashed line which divides area II in half. The resulting value "M" is referred to as the morphological index of the compound. Assuming that the line represents the locus of all cation combinations producing best fit of tetrahedral and octahedral sheets, the greater the value of M, the greater the misfit and therefore the greater the tendency of the layer to curve (other factors being equal). A value of +74.23* for the ideal chrysotile composition represents sufficient misfit to cause formation of tubes with "positive" curvature. Negative values indicate a tendency for the layers to curve in the opposite direction, as in the case of halloysite ($4H_2O$).

Analysis of the data reveals some interesting relationships. Cronstedtite lies directly on the line while (3) $Ge_4Mg_6O_{10}(OH)_8$, (4) $Ge_4Ni_6 O_{10}(OH)_8$, (5) $(Si_3Al)(Mg_5Al)O_{10}(OH)_8$, ferrous chamosite and amesite all lie within 40 units of it. The position of ferric chamosite (Brindley and Youell, 1953) was determined on the basis of the assumption that the oxidation of the ferrous to ferric iron produced no change in the distribution of the cations in the tetrahedral and octahedral sheets.

Since areas in Fig. 2 encompassing the kaolin and serpentine minerals fall in borderline situations with respect to fields I, II, and III, the amount of misfit produced by cation substitution leads to a sensitive situation such that the resultant morphology will be easily affected by and dependent upon the role of other factors such as interlayer bond strength, proportion of tetrahedral to octahedral cations and distribution of H^+. Nevertheless, the M values in Table VI reveal interesting differences between the minerals within each group.

In the serpentines the mean value for chrysotile is +71.94 as compared to +67.96 for all the platy varieties and +62.96 for material labeled antigorite. The fact that the highest value is given by one of the best examples of tubular chrysotile, and the lowest value is that of the megascopically platy, type antigorite, indicates that the method of evaluation is meaningful. The analyses of fiber (C-1 to C-4) and adjoining matrix (P-1(M) to P-4(M)) from the same specimens (Kalousek and Muttart, 1957) are of particular interest in that the difference in M values between the two portions of each sample is in the expected direction but very small. Thus, as might be expected in situations where matrix and fiber are intimately associated, the formation of one type rather than the other may hinge on differences in chemistry which are very slight and related to factors such as temperature, pressure, stress,

* The perpendicular distance to the line is calculated from the formula: $M = \sin 45°$ $(X - Y - .285)(1000)$.

TABLE VI. RADII OF TETRAHEDRAL AND OCTAHEDRAL CATIONS,
AND MORPHOLOGICAL INDEX: M

Sample	Average Cation Radius		M	Sample	Average Cation Radius		M
	IV	VI			IV	VI	
Ideal Serpentine	.390	.780	74.23	*Ideal Kaolinite*	.390	.570	−74.23
Chrysotile:				Dickite:			
C-1	.391	.779	72.82	D-1	.400	.572	−79.89
C-2	.391	.780	73.53	D-2	.396	.570	−78.48
C-3	.392	.779	72.11	D-3	.396	.570	−78.48
C-4	.392	.779	72.11	D-4	.391	.570	−74.94
C-5	.392	.779	72.11	D-5	.390	.570	−74.23
C-6	.391	.780	73.53	D-6	.390	.570	−74.23
C-7	.395	.778	69.29	D-7	.392	.572	−74.23
C-8	.396	.780	69.99				
				Kaolinite:			
Platy Serpentine:				K-5	.390	.572	−72.82
P-1(M)	.393	.779	71.40	K-6	.391	.573	−72.82
P-2(M)	.393	.778	70.70	K-7	.392	.571	−74.94
P-3(M)	.393	.779	71.40	K-8	.394	.571	−76.36
P-4(M)	.393	.779	71.40	K-9	.393	.571	−75.65
P-5	.403	.775	61.51	K-10	.396	.578	−72.82
P-6	.404	.774	60.09	K-11	.396	.571	−77.77
P-7	.402	.777	63.63	K-12	.396	.572	−77.06
P-8	.394	.778	69.99	K-13	.397	.571	−78.48
P-9	.394	.778	69.99	K-14	.397	.580	−72.11
P-10	.399	.778	66.46	K-15	.390	.571	−73.53
P-11	.397	.778	67.87	K-16	.390	.571	−73.53
P-12	.393	.779	71.40				
P-13	.393	.777	69.99	Halloysite:			
P-14	.405	.776	60.80	H-2	.390	.570	−74.23
P-15	.391	.779	72.82	H-3	.400	.570	−81.30
				H-4	.393	.570	−76.36
Other 1:1				H-5	.390	.571	−73.53
Am	.480	.715	−35.35	H-6	.394	.571	−76.36
Cr	.505	.789	−00.70	H-7	.394	.572	−75.65
Fe″-Ch	.456	.756	10.60	H-8	.399	.571	−79.89
Fe‴-Ch	.455	.655	−60.09	H-9	.392	.571	−74.94
				H-10	.393	.570	−76.36
Synthetic 1:1				H-11	.405	.571	−84.13
1	.390	.780	74.23	H-12	.395	.570	−77.77
2	.390	.780	74.23	H-13	.400	.572	−79.89
3	.440	.780	38.88	H-21	.392	.571	−74.94
4	.440	.780	38.88	H-22	.395	.570	−77.77
5	.435	.745	17.67	H-23	.397	.571	−78.48
				H-24	.393	.573	−74.23
Chlorite:				H-25	.399	.571	−79.89
Sh	.489	.722	−36.76	H-26	.394	.570	−77.06
Ri	.451	.749	9.19				
Ap	.456	.757	11.31	Group Averages:			
Th	.471	.751	− 3.53	Chrysotile	.393	.779	71.94
Cl	.441	.746	−14.14	Platy serpentine	.397	.778	67.96
Pe	.425	.756	32.52	Matrix (M)	.393	.779	71.23
Da	.476	.733	−19.80	Excluding (M)	.398	.777	66.78
P	.426	.782	50.20	Other 1:1	.474	.729	−21.39
Co	.453	.746	5.66	Chlorite	.454	.749	3.85
				Dickite	.394	.571	−76.35
				Kaolinite	.394	.573	−74.82
				Halloysite	.395	.571	−77.38

etc. which influenced crystallization at the time of formation or subsequent alteration of the rock.

In considering the significance of the M values, it is important to appreciate that most of the so-called platy varieties and massive specimens contain varying amounts of fibrous material. This has been demonstrated by Zussman, Brindley and Comer (1957) and by work by the author and Nagy as reported in the paper by Nagy and Faust (1956). Therefore, whereas the range in M values for the relatively homogeneous fibrous specimens is only from $+69.29$ to $+73.53$, that for the platy material is much greater ($+60.09$ to $+72.82$) due not only to a greater compositional range of the platy component but also to the probability that many of the analyzed specimens contained admixed, submicroscopic fibers which may approach the ideal composition. Until precise evaluations can be made of the chemical composition of individual submicroscopic plates, laths and tubes, it cannot be said which of these two factors plays the more important role.

In the kaolin group the range in M values is not quite as large as in the serpentines and the averages for kaolinites (-74.82), dickites (-76.35) and halloysites (-77.38) are not greatly different. Since, with one or two exceptions, the role of Mg is minor, the variation in M values is a result of the variation in the amount of Al and Fe''' in excess of Si.

The M values indicate that, although the difference for platy (kaolinite and dickite) versus lath-shaped and tubular varieties (halloysite) is in the right direction, variation in misfit due to cation distribution in octahedral and tetrahedral sheets is probably not the major cause of variation in morphology within the group.

Morphology in Relation to Proportions of Major Components

In order to show the relationships of bulk composition to morphology each analysis was recalculated so that the sum of the major components would be equal to 100. The resulting compositions are plotted on Figs. 3 and 4. It is apparent in the case of both the serpentine and kaolin groups that the chief difference between platy and tubular varieties lies in the greater amount of hydrogen and associated oxygen in the latter.

In the serpentines (Fig. 3) the three components evaluated are SiO_2, H_2O+ and R''O where R'' = Mg, Fe'', Mn and Ni. If the assumption is correct that Al and Fe''' are divided equally between tetrahedral sites (substitution for Si) and octahedral sites (substitution for R''), the effect of these ions would be to move each point to the left in a direction parallel to the dotted line which, if extended, would terminate at the H_2O corner of the diagram. Since, as shown in Fig. 2, the platy varieties contain more Al and Fe''' than chrysotile, the result of including these

components would be to increase the separation of the two groups.

As in the case of the data recorded in Table VI, the samples of adjoining matrix and fiber described by Kalousek and Muttart are very similar in composition suggesting that 1) the matrix is a mixture containing a large percentage of fiber, 2) the platy component of the matrix has a composition very similar to the fiber, 3) or both factors are involved.

Of the remaining eleven platy samples, eight differ markedly from the chrysotiles by having a greater $SiO_2:R''O$ ratio. It is significant in this respect, that the greatest departure from the ideal serpentine composition is made by varieties labeled deweylite, williamsite, baltimoreite and Yu Yen Stone; whereas analyses of material labeled antigorite (Nos. 5, 6, 7, 8, 9) plot closer to the line which indicates the ideal proportion of the components in question. The "anomalous" character of sample P-10 (one of two antigorites from Nikka Vord quarry, Shetland Islands) has been discussed previously by Brindley and von Knorring (1954), McConnell (1954) and Zussman (1956).

The relationships of composition and structure of sample P-9 have been considered by Hess, *et al.* (1952) and Brindley (1954); and of samples P-8 and P-9 by Zussman (1954).

The relationships shown diagrammatically in Fig. 3 indicate that on the whole, platy crystals are favored over fibrous material if there is 1)
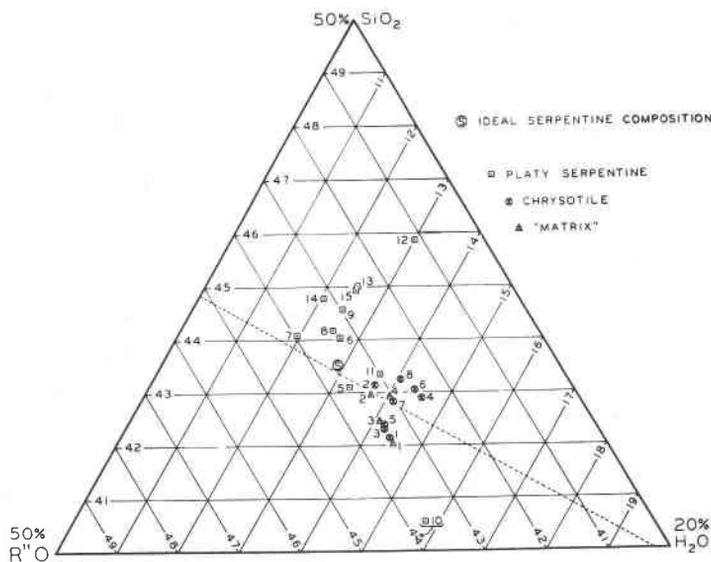
FIG. 3. SiO_2-(Mg, Fe'', Ni, Mn)O-H_2O composition of serpentine samples.

an increase of SiO_2 with respect to $R''O$, and 2) a decrease in H_2O+. The role of H^+ will be considered later. If the assumption is correct that Al and Fe''' are equally divided between tetrahedral and octahedral positions, the evidence with respect to $SiO_2:R''O$ ratio is in line with the observation made earlier that the amount of misfit is affected by the number of ions in the tetrahedral sheet relative to a given number in the octahedral sheet. In this case, the increase in proportion of Si to divalent ions will improve the fit between the larger "ideal" brucite sheet and smaller "ideal" Si-O sheet and thereby promote the formation of platy crystals.

In the kaolin group (Fig. 4) the range in the ratio of $SiO_2:Al_2O_3$ shown by kaolinites and dickites is similar to that shown by most of the halloysite samples. However, as indicated by the work of others, the average $SiO_2:Al_2O_3$ ratio of halloysites is lower than that of kaolinites and dickites, being in this case 192:100 (excluding samples 3, 11 and 26) as compared with 196:100. Granting that the difference may be due to undetectable impurities in the samples analyzed, the fact remains that the lath-shaped and tubular material has 1) a lower $SiO_2:Al_2O_3$ ratio and 2) a higher H_2O+ content than the platy varieties. It is important to bear in mind that most if not all of the interlayer water found in halloysite ($4H_2O$) is presumably not included in the H_2O+ value of the analysis.
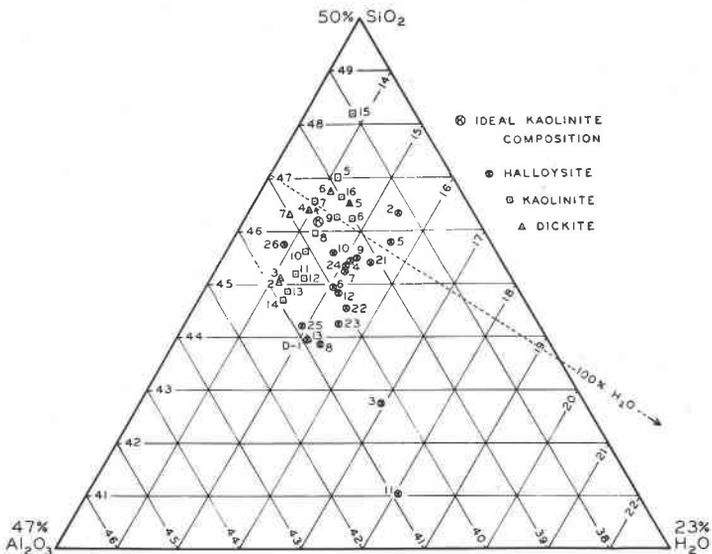
Fig. 4. SiO_2-Al_2O_3-H_2O composition of kaolin samples.

OTHER PERTINENT RELATIONSHIPS

General Statement

The morphological-chemical relationships discussed thus far are considered to be particularly significant because they involve a minimum number of assumptions. Thus, the general relationships graphed in Figs. 2, 3, and 4 pertain regardless of the choice of radii used to represent the ions, of the means of proportioning of cations between tetrahedral and octahedral positions, and of the problem as to the exact disposition of hydrogen. The writer, therefore, wishes to draw a sharp distinction between the previous discussion and the points made in this section in that the latter rely more directly upon the assumptions used in making the structural calculations. Although these points are more subject to question, the calculations have resulted in a number of interesting relationships which, in the opinion of the author, should be considered by other workers in this area. Two things, in particular, deserve attention. These are: 1) the number of cations (excluding H^+) assigned to tetrahedral as opposed to octahedral positions; and 2) the amount of H_2O+ given in the analysis as compared with that needed to provide the "proper" proportion of (OH) and oxygen with respect to the postulated cation distribution in the formulas.

Number of Tetrahedral versus Octahedral Cations

Table VII lists the pertinent cation distribution data for all minerals except those of the kaolin group. These are not included because the method of allocating ions to tetrahedral versus octahedral positions in the formulas for this group results in equal distribution except in those few cases where appreciable divalent ions are present. Column 1 gives the number of cations allocated to tetrahedral positions, column 2 those in octahedral positions, and column 3 the number of tetrahedral ions on the basis of six in the octahedral sheet.

In the serpentines the distribution corroborates the evidence presented in Fig. 3 to the effect that, assuming equal amounts of Al and Fe''' in the two sheets of each layer, there are appreciably more tetrahedral cations per six octahedral cations in the platy than in the fibrous specimens. Thus, the average chrysotile has 4.041 tetrahedral ions for every six octahedral ions whereas in the platy serpentines (omitting the matrix specimens and the anomalous specimen from Unst, P-10) the ratio is 4.291:6.0. If correct, this would require nearly one extra tetrahedral position for every three octahedral units of structure* in the layer. Ob-

* Using a formula unit with six octahedral positions.

TABLE VII. CATION DISTRIBUTION DATA

Sample	No. of Cations: IV	No. of Cations: VI	Cat$_{IV}$ per Cat$_{VI}$ =6	Sample	No. of Cations: IV	No. of Cations: VI	Cat$_{IV}$ per Cat$_{VI}$ =6
Chrysotile:				Other 1:1:			
C-1	3.863	5.903	3.926	Am	3.980	5.896	4.050
C-2	3.955	5.885	4.032	Cr	3.700	6.141	3.615
C-3	3.884	5.916	3.939	Fe″-Ch	4.184	5.658	4.437
C-4	3.921	5.928	3.969	Fe‴-Ch	4.435	6.154	4.324
C-5	3.942	5.747	4.116				
C-6	3.936	5.780	4.086	Chlorite:			
C-7	3.947	5.853	4.046	Sh	4.244	5.950	4.280
C-8	4.012	5.715	4.212	Ri	4.028	5.983	4.039
				Ap	4.035	5.795	4.178
Platy Serpentine:				Th	4.410	5.486	4.823
P-1(M)	3.853	5.901	3.918	Cl	4.051	5.873	4.139
P-2(M)	3.948	5.957	3.976	Pe	4.003	5.805	4.138
P-3(M)	3.907	5.926	3.956	Da	4.348	5.463	4.775
P-4(M)	3.920	5.828	4.036				
P-5	4.098	5.850	4.203	Group Averages:			
P-6	4.176	5.794	4.324	Chrysotile	3.933	5.841	4.041
P-7	4.243	5.883	4.327	Platy serpentine	4.075	5.868	4.169
P-8	4.123	5.939	4.165	Excluding (M)			
P-9	4.171	5.885	4.253	and P-10	4.180	5.846	4.291
P-10	3.698	5.940	3.735	Chlorite	4.160	5.765	4.339
P-11	4.021	5.864	4.114				
P-12	4.232	5.592	4.541				
P-13	4.170	5.857	4.265				
P-14	4.406	5.946	4.446				
P-15	4.163	5.843	4.275				

viously this would be one way in which the misfit between the smaller Si-O and larger "brucite" sheets might be partially adjusted.

Although the presence of more than four filled tetrahedral positions for every six in the octahedral sheet involves a departure from the "classical" picture of the serpentine structure, it is in accord with the structural data presented by Zussman (1954) as evidence in support of an alternating or rectified wave structure for some varieties of platy serpentine. The cation assignments he gives as the result of detailed structural analysis of antigorites from Venezuela and New Zealand are compared in Table VIII with those calculated by the author from the same analyses. The ratios obtained for the number of tetrahedral ions per six octahedral ions are given at the bottom of the table. The close agreement indicates that the method used herein for calculating the formulas from the analyses is meaningful.

Amesite and ferrous and ferric chamosite also have ratios greater than 4:6 whereas cronstedtite gives a value of 3.6:6.0.

As in the other evaluations the matrix and fiber pairs give very similar values.

TABLE VIII. STRUCTURAL ASSIGNMENTS FOR NEW ZEALAND AND VENEZUELA
ANTIGORITES (P-8 AND P-9)

Ions		Ideal structure*	Mikonui, New Zealand		Caracas, Venezuela	
			Zussman	calculated	Zussman	calculated
IV	O	48.9	51.0	50.2	51.0	50.7
	Si, etc.	32.6	33.9	33.6	34.1	34.0
	O	32.6	34.4	33.0	33.9	33.0
VI	(OH)	16.3	15.0	16.2	15.0	16.1
	Mg, etc.	48.9	48.9	48.4	48.2	48.0
	(OH)	48.9	47.6	48.7	47.4	48.3
Cat.$_{IV}$ per Cat.$_{VI}$ = 6		4.00	4.16	4.16	4.24	4.25

* Proportioned to Zussman's structural unit of 8.15 "cells" along "a" axis.

Excess or Deficiency of Hydrogen and Oxygen

With respect to item 2 of the preceding "general statement," it is of interest that the amount of oxygen and hydrogen provided by each analysis is not equal to that needed if these elements are proportioned with respect to the cation distribution in octahedral and tetrahedral sheets as indicated by the ideal structural formula.

Thus, in the kaolinite example given in Table II, 0.052 more oxygen ion and 0.105 more H^+ is needed for the properly proportioned structural formula than the H_2O+ of the analysis provides. In the serpentine example, on the other hand, the analysis provides an excess of 0.483 oxygen and 0.965 H^+ over that needed in a "properly proportioned" structure. Although in writing the structural formulas in Table V it is convenient to add "excess" hydrogen to the tetrahedral sheet as H_4 or to account for a deficiency by subtracting the element (with appropriate oxygen) from the octahedral sheet, the author feels that correct assignments must await procurement of better experimental data. The following discussion, therefore, relates only to the amount of these ions present and not to the possible structural positions. The data are presented in the last two columns of Table IV. For each analysis the amount of excess or deficient hydrogen should be twice that of the corresponding oxygen. The slight discrepancies are due to rounding-off errors in the calculations.

Because of the uncertainty as to the temperature at which H_2O+ was measured for many of the analyses, the precision of the values is uncertain. However, an analysis of variance shows that the differences between chrysotiles as opposed to platy serpentines (including matrix

samples), and between kaolinites and dickites as opposed to halloysites are significant at the 1% level. The chrysotiles have a mean value of $+.821$ H^+ as compared to $+.125$ for platy serpentines, and halloysites give a mean of $+.889$ compared to $+.099$ for kaolinites and dickites.

Again it will be noted that the values for the "matrix" analyses from the matrix and fiber pairs (numbers P-1(M) to P-4(M)) are very different from those of the platy serpentines and very similar to those of the adjacent fiber material. The anomalous position of sample P-10 from Unst is apparent.

Data on Chlorites

The relationship of serpentine to chlorite has been discussed by many workers, among the most recent being Nelson and Roy (1954), Brindley and Gillery (1954 and 1956), Bradley (1955) and Gillery (1958). The purpose of including the subject in this paper is to point out that morphological and other relationships between the two groups of minerals can be satisfactorily explained on the basis of the same parameters as those used to evaluate the 1:1 layer minerals. The pertinent data are presented in this section and will be discussed in the next.

A number of chlorite analyses considered by Orcel (1927) to be characteristic of various chlorite sub-groups have been evaluated on the same basis as the other analyses considered here, and the various compositions are represented on the graph in Fig. 2. In addition characteristic compositions of corundophyllite and penninite are also represented, as is that of the daphnite specimen discussed by Brindley and Gillery (1954). It is apparent that the chlorites represented fall well within area II and do not overlap the serpentine compositions. The relative positions are brought out by the M values in Table VI. Those for chlorite compositions range from -36.76 for sheridanite to $+50.20$ for penninite. The lowest value for a platy serpentine is $+60.09$ for the Val Antigorio antigorite.

It is also instructive to consider the amount of hydrogen present in the H_2O+ of the analysis as compared with that "required" to provide the proper proportion of OH as given by the ideal chlorite formula. The values are recorded in Table IV (the amounts given are for one half of the usual formula unit in order that the values may be more easily compared with those of the 1:1 minerals). In marked contrast to the chrysotiles and all but one of the platy serpentines, the chlorite analyses have appreciably less hydrogen than that needed if the proportions are to be similar to those in the formula.

DISCUSSION

The Role of Hydrogen

In those minerals, such as amesite, cronstedtite, chamosite, platy serpentines and chlorites, where the analysis provides less H^+ and $O^=$ than is appropriate to the ideal formula, it is apparent that the lattice must contain oxygens in place of some of the hydroxyls and either have an occasional oxygen or hydroxyl position vacant or have a structural modification, such as the rectified layers postulated by Zussman (1954), which involves fewer oxygen and hydroxyl positions than the ideal structure.

In the opposite situation, namely in hallosysite and chrysotile, where the analysis provides an excess of hydrogen and oxygen, either 1) some interlayer water is so tightly held as to be included in the H_2O+ value, or 2) the ions occupy structural positions in the 1:1 lattice. Independent of whether the hydrogen ions are present between the layers in H_2O or $(H_3O)^+$ or are in the tetrahedral sheet as OH in place of or in addition to oxygen, a very probable and important result of their presence is a weakening of interlayer bonds. This is evidenced by the difference in the (001) spacings of halloysite (7.3–7.9 Å) as compared with kaolinite (7.15 Å) and dickite (7.15 Å), and of chrysotile (7.3 Å) as compared with platy serpentine (7.26 Å).

Interrelationships of Chemistry, Structure and Morphology

Some of the relationships between the mineral groups with respect to the parameters discussed herein are shown in Fig. 5. The figures in parentheses give the (001) and a_o values for the various structures. Some observations of a general nature are as follows:

1) The halloysites, kaolinites and dickites, chrysotiles, and platy serpentines fall in distinct groups. The matrix specimens closely associated with serpentine fiber fall within the chrysotile area as in Fig. 3. Platy serpentine P-10 and halloysite H-26 are anomalous.

2) With respect to both "amount of misfit" and "excess or deficient hydrogen" the analogy between platy serpentine and kaolinite and dickite, and between chrysotile and halloysite is very apparent.

3) The compounds with the greatest amount of misfit have the largest amount of H^+ and the smallest amount of substitution of other cations. These compounds crystallize in the form of tubes or curved laths.

4) The gap between kaolins and serpentines is bridged, in the lower half of the diagram only, by the other 1:1 minerals and the chlorites.

5) On the left and right sides of the diagram, the (001) spacing characteristic of the minerals of each group increases with increasing H^+ content.

6) The a_o spacing increases from that of the kaolins to a maximum for cronstedtite at the center of the diagram and then decreases again to that of chrysotile.
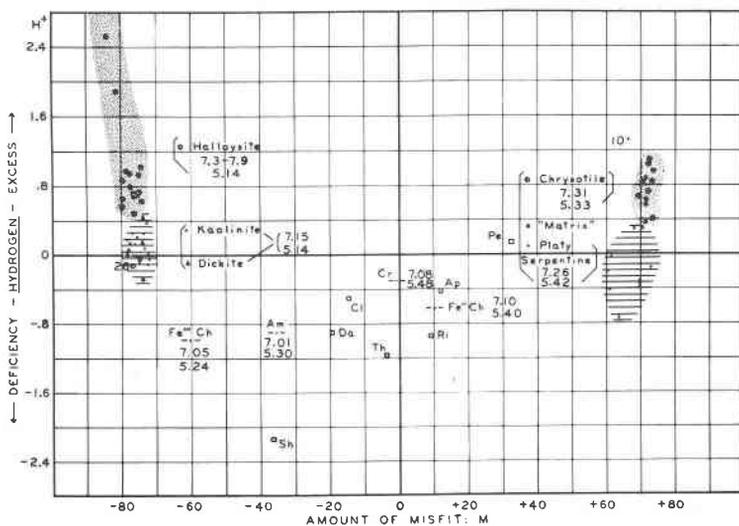
Fig. 5. Relationship of excess or deficient hydrogen content to the amount of misfit of tetrahedral and octahedral sheets. The top number is the (001) spacing whereas the bottom number is a_o.

7) Platy crystals are either deficient in H^+ or have enough to approach the correct proportions of the structural formula. Theoretically, compounds having the largest platy crystals have the least amount of misfit and fall nearest the center of the diagram.

These relationships all support the contention that, in this group of the sheet structure silicates, details of structure and morphology are a function of the amount of misfit and the strength of interlayer bonds. These latter factors are interdependent and are a function of chemical composition. Interlayer bonds are strongest in amesite, cronstedtite, chamosite and the chlorites where excess H^+ is not present and where excess negative charges produced by substitution in the tetrahedral sheet form bonds not only with positive charges produced by substitution in the octahedral sheet of the same layer, but also with those produced in the neighboring octahedral sheet in the adjacent layer.

Toward the left side of the diagram, as the amount of substitution decreases, the role of hydroxyl bonds becomes more important. In kaolinite and dickite these bonds are presumably strong enough to "stretch" the gibbsite sheet of one layer to fit the Si-O sheet of the adjacent layer thereby overcoming the misfit and producing platy crystals of limited size. Oberlin and Tchoubar (1957) have shown that when thin enough plates of kaolinite are produced either naturally or synthetically, interlayer bonds are weakened and the plates curl.

With an increase in excess H^+ above that found in kaolinites and dickites, interlayer bonds become too weak to overcome the misfit and curved laths and tubes are to be expected. Where excess H^+ is sufficient in amount ($= +6.546$) to combine with associated oxygen in a continuous sheet of interlayer water, tubes of halloysite ($4H_2O$) are formed. For the specimens shown in Fig. 5 only H_2O+ was considered and consequently only compositions approaching halloysite ($2H_2O$) are represented. Although it was formerly believed that all $2H_2O$ halloysite probably formed by dehydration of the $4H_2O$ material, present evidence (Bates and Comer, 1959) indicates that $2H_2O$ halloysite such as that pictured in Fig. 1 may form directly where there is enough excess H^+ to cause sufficiently weak interlayer bonds for the production of curved laths but not enough to make the bonds so weak that tubes result.

Toward the right side of Fig. 5 the situation differs from that just discussed in that hydroxyl bonds are formed only where Al and Fe''' substitute for Mg. In the platy serpentines, as compared with chrysotile, the decrease in amount of misfit and the OH bond formation produced by Al and Fe''' substitution is sufficient to produce crystals which are characteristically platy but show irregular rather than crystallographic outlines. However, interlayer bonds are significantly weaker than in kaolinite and it is not surprising that plates may exist that may be made up of alternating or rectified waves of varying periodicity giving rise to varying a_o dimensions (Zussman, Brindley and Comer, 1957).

In chrysotile, with practically no Al and Fe''' substitution and with a large amount of excess H^+, interlayer bonds are so weak and the misfit so great that tubes are the logical result.

Finally, the minerals lying between the kaolin and serpentine groups have the maximum amount of Al and Fe''' substitution and therefore 1) relatively strong interlayer bonds, 2) the least amount of misfit of octahedral and tetrahedral sheets, 3) greater order perpendicular to (001) and consequently 4) larger platy crystals with crystallographic outlines.

Summary

Because of their 1:1 layer structure the kaolin and serpentine groups, and certain other structurally associated minerals and synthetic compounds, have morphological and structural characteristics which are particularly sensitive to slight changes in chemical composition. In order to demonstrate the nature of the interrelationships of chemistry, structure and morphology and their bearing on similarities and differences within and between the mineral groups, 64 analyses have been evaluated and compared.

The evaluation has been made in terms of four parameters: 1) a

morphological index, M, which is a measure of the amount of misfit of tetrahedral and octahedral sheets; (2) the proportions of the major components, SiO_2, Al_2O_3 (or $R''O$) and H_2O, as represented on ternary diagrams; 3) the number of cations assigned to the tetrahedral sheet in relation to a standard number in the octahedral sheet; and 4) the amount of deficient or excess hydrogen and oxygen recorded as H_2O+ in the chemical analysis as related to that required to provide the correct number of properly proportioned O and OH ions in the structural formulas.

Conclusions resulting from the use of parameters one and two are considered particularly significant because they do not depend on the validity of assumptions that must be made in the calculation of structural formulas. Parameters three and four involve the assumption that the Al and Fe''' in excess of the amount of silicon in the dioctahedral minerals and all Al and Fe''' in the trioctahedral minerals is divided equally between the tetrahedral and octahedral sheets.

The use of these parameters helps to clarify the differences within and between mineral groups. With respect to the overall group of 1:1 minerals, the two major chemical differences are: 1) the nature and number of cations substituting for Si in the tetrahedral sheet and for Al or Mg in the octahedral sheet; and 2) the amount of H_2O+.

Variation in the amount of misfit of octahedral and tetrahedral sheets brings out not only the expected distinction among the kaolin group, the serpentine group, and the other 1:1 minerals (amesite, cronstedtite and chamosite) but also reveals a smaller but distinct difference between chrysotiles and platy serpentines. It is of interest that the difference between chrysotile and the type antigorite from Val Antigorio is greater than that between chrysotile and any other platy serpentine. Of the other 1:1 minerals cronstedtite has the least amount of misfit and all three have much less than kaolin minerals or serpentines. The morphological and misfit relationships of several synthetic silicates and germanates fit nicely in the pattern set by the natural materials.

The amount of H_2O+ varies considerably in the analyses studied and serves particularly well to distinguish platy from lath-shaped and tubular varieties in the kaolin and serpentine groups since analyses of platy minerals contain significantly less H_2O+ than those of the non-platy specimens.

Calculation of structural formulas is based on a structural unit of 18 oxygens, but the distribution of the oxygens is determined not in terms of an ideal structural formula but on the basis of the distribution of cations in the two sheets of the layer. This approach is used in the belief that the number of cations (and accompanying oxygens) in the tetra-

hedral sheet, per standard number in the octahedral sheet, may vary in order to reduce the amount of misfit between sheets. Thus, whereas the chrysotiles have, on the average, 4.041 tetrahedral cations for every six octahedral cations the ratio in the platy serpentines (excluding anomalous sample P-10 and matrix samples) is 4.291. Obviously, this excess could increase the size of the tetrahedral sheet and thereby produce a better fit with the larger "brucite" sheet.

The method of formula calculation also provides a measure of the amount of excess or deficient hydrogen and oxygen in the H_2O+ value of the chemical analysis as related to that required by properly proportioning O and OH in accord with the formula distribution of cations other than hydrogen. The results bear out the bulk-composition evidence to the effect that tubular and lath-shaped varieties in both kaolin and serpentine groups have a significant excess of hydrogen and oxygen in the H_2O+ value of the analysis as compared to that "required" by the cations. On the other hand platy varieties of these groups have either a deficiency or nearly the correct amount of H_2O+, and the other 1:1 minerals have a deficiency.

Since H_2O+ content, which bears directly on interlayer bond strength, and cation nature and distribution in the two sheets of each layer are probably interrelated, both factors must be considered in explaining structural and morphological details. Figure 5 represents an attempt to do this and thus place the individual specimens and various groups in proper perspective relative to one another. When viewed in this way distinctions between groups become sharply defined and relationships between morphology and chemistry become more apparent. The picture also serves to suggest that the transition from kaolinite to halloysite ($2H_2O$) is simply the first part of the series leading to halloysite ($4H_2O$) and allophane as H_2O content is increased. In the serpentines the difference between chrysotile and the platy varieties is distinct but the variation within the platy group is probably due in part to mechanical mixtures of fiber and platy material in the samples analyzed.

A few typical chlorite analyses have been included to demonstrate the relationship of these minerals to the 1:1 group. On the basis of the parameters used herein, the chlorites characteristically have a deficiency in the H_2O+ content of the analyses and show a much smaller "amount of misfit" than serpentines or kaolins. In general they have similar characteristics to the 1:1 minerals amestie, chamosite and cronstedtite thus indicating the very close chemical and structural relationships of the two mineral groups. The fact that there is no overlap of chlorites and serpentines is logical in that a chlorite structure would be expected only

where there is sufficient substitution of trivalent ions for Si and Mg to keep the amount of misfit small and to promote the formation of relatively strong interlayer bonds.

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