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CHEMICAL COMPOSITION AND GENESIS OF GLAUCONITE AND CELADONITE*

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ABSTRACT

A method of study found applicable to the clay minerals clarifies the chemical relations of glauconite and celadonite, and gives significant information about their genesis.

The representative formula of glauconite is:

$$(K,\ Ca_{\frac{1}{2}},\ Na)_{.84}(Al_{.47}Fe'''_{.97}Fe''_{.19}Mg_{.40})(Si_{3.65}Al_{.35})O_{10}(OH)_2$$

and of celadonite is:

$$(K,\ Ca_{\frac{1}{2}},\ Na)_{.84}(Al_{.34}Fe'''_{.76}Fe''_{.24}Mg_{.76})(Si_{3.89}Al_{.11})O_{10}(OH)_2,$$

which represent very closely compositions of a large number of mineral specimens. These formulas show that both minerals belong to the heptaphyllite group of micas.

A supply of ferrous iron (necessitating a reducing environment), magnesium, and potassium, is a controlling condition for the formation of both minerals. Glauconite, which forms only in a marine environment, maintained in a reducing condition by bacterial action, derives its magnesium and potassium from the sea water, and its other constituents from mud. Celadonite, which commonly forms in vesicular basalts, derives its essential magnesium, iron, and silica from olivine, and its other constituents from deuteric solutions.

INTRODUCTION

In the course of work on the montmorillonite-nontronite group of clay minerals it became evident that the methods used in correlating chemical analyses of that group might give interesting information on other minerals related to the micas. Glauconite being a mica is a mineral of this type, and its widespread occurrence has caused it to be the subject of many studies. Glauconite has formed as a marine deposit in every geological age since the pre-Cambrian and is found in many modern marine sediments. A material known as celadonite that occurs as vesicular fillings in basaltic rocks, has been correlated with glauconite.[1,2] Glauconite has been

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* Approved for publication by the Chief of Bureau of Plant Industry and Director of U. S. Geological Survey.

[1] Lacroix, A., *Mineralogie de la France et de ses Colonies,* **1,** part 1, 406–409 (1893).

[2] Glinka, K., Der Glauconite, sein Entstehung, sein chemischer Bestand, und die Art und Weise seiner Verwiterung: *Pub. de l'Institut agromique de Novo. Alex Russie, St. Petersberg* **(1896)**.

identified as a mica by several investigators,[3,4,5,6,7] and celadonite, although less widely investigated,has been assigned to the micas by Glinka and this has been verified by Maegdefrau and Hofmann[8] using x-ray diffraction methods, and confirmed by our own observations.

These calculations of the chemical relations of glauconite and celadonite are based on the assumption that there are no major analytical errors in the analyses, and that the material analyzed was fairly free from impurities. It is believed that the consideration of an adequate number of analyses will obviate the difficulty introduced by minor errors and that major ones will become self evident. The method of consideration is based on the establishment of a consistent trend in composition that agrees with crystal structure, and recognized ionic substitutions. Measured by these criteria, the available analyses seem to give a consistent picture; in fact, the departures from this trend are less important than were to be expected.

MODE OF OCCURRENCE AND PHYSICAL PPOPERTIES

Glauconite as it occurs in sedimentary rocks is so well known and has been so often described that little need be said about its occurrence and properties.

Sedimentary glauconite most commonly occurs as rounded pellets from one to several millimeters in diameter, its formation by replacement of foraminifera being on the whole exceptional. It is commonly made up of an exceedingly fine-grained, but obviously crystalline aggregate of overlapping crystal plates. However, almost all who have investigated glauconite[9,10,11,12,13,14,15] have found grains that were crystal units.

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[3] Murray, Sir John, *Challenger Rept., Deep sea deposits*, 239 (**1891**).

[4] Lacroix, A., *op. cit.*

[5] Glinka, K., *op. cit.*

[6] Schneider, H., A study of glauconite: *Jour. Geol.*, **35,** 289–310 (1927).

[7] Gruner, John W., The structural relationship of glauconite and mica: *Am. Mineral.*, **20,** 699–714 (1935).

[8] Maegdefrau, E., and Hofmann, U., Glimmerartige Mineralien als Tonsubstanzen: *Zeits. Krist.*, **A98,** 31–59 (1937).

[9] Lacroix, A., *op. cit.*

[10] Cayeux, L., *Contribution a l'etude des Terrains Sedimentaires, Lille*, 163–164 (**1897**).

[11] Mansfield, G. R., Potash in the greensands of New Jersey: *U. S. Geol. Surv., Bull.* **727,** Plate, 4D (1922).

[12] Hadding, A., The pre-Quaternary sedimentary rocks of Sweden. IV. Glauconite and glauconitic rocks: *Medd. Lunds Geol.-Mineral Institute*, **51,** 51 (1932).

[13] Ross, Clarence S., The optical properties and chemical composition of glauconite: *Proc. U. S. Nat. Museum*, No. 2628, **69,** 1–15 (1926).

[14] Schneider, Hyrum, *op. cit.*

[15] Ichimura, T., Some glauconitic rocks from Taiwan (Formosa): *Memoirs Faculty Science and Agri., Taihoku Imperial Univ.*, **22,** No. 3, 25–63, plate 4 (1940).

Some of these are roughly hexagonal in outline, and Lacroix, Cayeux, and Ross, found evidence of a small inclination of the optic axis, thus indicating monoclinic symmetry. The optical properties reported agree within fairly narrow limits and those determined (Table 1) on the unusually good material from Bonne Terre, Missouri,[16] are adequately representative.

The properties of celadonite have not been determined as fully as have those of sedimentary glauconite, and so the occurrence and properties will be described in the following section.

The name celadonite was proposed by Glocker[17] in 1847, although the same material had been previously described as terra verte[18] and grünerde.[19] Celadonite is most abundant in vesicular cavities in basalt; but it also replaces olivine, and less commonly hypersthene or groundmass material. It forms radial or vermicular aggregates that have commonly been described as fibrous, but detailed study indicates that it is made up of minute elongated plates, and that the habit is bladed. In hand specimens the appearance is earthy, but in good light reflections from minute crystal faces may be observed. The color is commonly blue-green in the hand specimen, and under the microscope the material commonly has a decided blue cast in the direction of maximum absorption, and is yellow, yellow-green, or pale green in the other direction. The mode of occurrence of representative samples of the celadonite are as follows:

The material from Reno, Nevada, occurs as vesicular fillings in a fine-grained olivine basalt. The olivine has locally been partly altered to the same material, but in general there has been complete alteration to serpentine. The filled vesicular cavities in the basalt vary in size up to a diameter of 12 millimeters, and thus are unusually large. The color of the compact material is light Danube green (Ridgway) and the powdered material Montpelier green. Most of the cavities are completely filled with celadonite, but a few are hollow or contain irregular areas of iron-rich saponite (as shown in Plate 1, Fig. *d*). A few of the smallest cavities contain only saponite.

The material from Sandoval County, New Mexico (collected by B. C. Renick), a photomicrograph of which is shown as Plate 1, Fig. c, fills more or less irregular or flattened vesicular cavities that reach a maximum length of 10 millimeters. The color in the hand specimen is methyl green (Ridgway). Some of the vesicles contain only celadonite, but others contain associated calcite and saponite.

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[16] Ross, Clarence S., *op. cit.*, p. 3.

[17] Glocker, E. F., Generum et Specierum Mineralium secundum Ordines Naturales digestorum Synopsis, p. 193, *Halle* (**1847**).

[18] De Lish, Romé, Cristallographie, ou Description des formes propres à tous les corps du Regne mineral, **2,** 502, Paris (1783).

[19] *Hoffmann-Bergmannisches Jour.*, 519 (**1788**).

Plate 1

Glauconite and celadonite

Fig. (*a*). Altered volcanic materials, Center Point, Arkansas. Dark grains in upper two-thirds of figure are phonolite rock fragments almost completely altered to glauconite. Large grain in lower one-third is augite replaced by glauconite. Light gray interstitial material is calcite. ×54.

Fig. (*b*). Large central portion is a phenocryst of olivine in basalt; nearly fresh in lower portion, but upper part (gray) has been replaced by celadonite. White areas, plagioclase; black, groundmass. Locality unknown. ×54.

FIG. (*c*). Celadonite filling of cavity in basalt, Sandoval County, New Mexico. Radial, bladed habit is typical of celadonite. ×54.

FIG. (*d*). Celadonite filling vesicle in basalt, Reno, Nevada. Gray portion celadonite, nearly white areas are saponite. ×40.

Optical properties of these samples and of one from Kern County, California, are given in Table 1. The *x*-ray powder diffraction patterns of these samples of celadonite and of glauconite showed no observable differences.

TABLE 1. OPTICAL PROPERTIES OF CELADONITE AND GLAUCONITE

Locality	Indices of refraction			Birefringence $\gamma-\alpha$	Optical character and 2V	Pleochroism[a]	
	α	β	γ			X	Y and Z
Celadonite							
Reno, Nev.	1.610		1.641	.031	—	Pale yellow green	Guinea green
Sandoval Co., N. Mexico	1.606		1.634	.028	—	Dull yellow green	Skobeloff green
Red Rock Canyon, Kern Co., Calif.	1.606		1.635	.029	—	Light yellow green	Ethyl green
Glauconite†							
Bonne Terre, Mo.	1.597	1.618	1.619	.022[b]	−20°	Lemon yellow	Dark Russian-green

[a] Color names following Ridgway.
† Ross, Clarence S., *op. cit.*
[b] $r>v$ distinct.

METHOD OF CALCULATING FORMULAS FOR MINERALS HAVING SILICATE LAYER LATTICES

The significance of the kind and extent of isomorphous replacements in the micas was first recognized by Maugin.[20] Elucidation of the general structural scheme of mica is due to Pauling[21] who further showed that a

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[20] Maugin, C., Étude des micas au moyen des rayons X: *Compt. Rend.*, **186,** 879–881, 1131–1133 (1928).

[21] Pauling, L., The structure of the micas and related minerals: *Proc. Nat. Acad. Sci.*, **16,** 123–129 (1930).

large group of minerals have related structures. Among these are micas, brittle micas, chlorites, vermiculites, stilpnomelanes, hydrous micas, pyrophyllite, talc, montmorillonite, beidellite, nontronite, saponite, cronstedite, glauconite, celadonite, and the kaolin minerals.

The mica-like minerals have the common structural feature of tetrahedral groups of oxygen ions about silicon ions, joined into a hexagonal layer by sharing of oxygen ions by two tetrahedra. Two such layers are joined by octahedral coordination about Al''', Mg'', etc., of oxygen ions that are not shared between tetrahedra. The potassium ions fit between two such composite layers and have twelve neighboring oxygen ions.

The succession of atomic layers along the normal to the micaceous cleavage of muscovite and phlogopite and their idealized chemical formulas are:

	Muscovite		*Phlogopite*
Interlayer Nx	K^+	Nx	K^+
	$6O^{--}$		$6O^{--}$
Tetrahedral	$1Al^{+++}3Si^{++++}$	$1Al^{+++}3Si^{++++}$	
	$4O^{--}2(OH^-, F^-)$		$4O^{--}2(OH^-, F^-)$
Octahedral	$4Al^{+++}$		$6Mg^{++}$
	$4O^{--}2(OH^-, F^-)$		$4O^{--}2(OH^-, F^-$
Tetrahedral	$1Al^{+++}3Si^{++++}$		$1Al^{+++}3Si^{++++}$
	$6O^{--}$		$6O^{--}$
Interlayer	K^+		K^+

Muscovite $K \cdot (Al_2) \cdot (Al, Si_3) \cdot O_{10} \cdot (OH, F)_2$
Phlogopite $K \cdot (Mg_3) \cdot (Al, Si_3) \cdot O_{10}(OH, F)_2$

General knowledge of silicate structures[22] leads to the following expected types of isomorphous replacements in the micas:

Interlayer positions	K^+, Na^+, Rb^+, Cs^+, Ca^{++}, Ba^{++}, Sr^{++}
Tetrahedral positions	Si^{++++}, Al^{+++}
Octahedral positions	Al^{+++}, Ti^{++++}, Fe^{+++}, Fe^{++}, Mn^{++}, Cr^{+++}, Mg^{++}, Li^+

A remaining question to be answered for the micas is the actual extent to which various replacements take place.

In the montmorillonite-nontronite group of minerals, and in the hydrous micas, the interlayer positions are not completely filled, and the present study indicates that this is also true for glauconite and celadonite. The analyses of glauconite and celadonite show a potash ratio varying from 1, the value normal for micas, down to 0.56, with but 7 below 0.75; the average being 0.842. Thus the deviation from complete filling of the interlayer positions is significant, but not excessive. This must be taken into account in the formula which will be of the general type X_A(Al, Fe'''.

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[22] Bragg, W. L., *Atomic Structure of Minerals*, Ithaca (**1937**).

Fe'', etc.$)_\Sigma(Al, Si)_4O_{10}(OH, F)_2$, where A is 1.00 or less and Σ is the number of octahedral positions filled, which must be 3.0 or less. The equivalence of the external base X is determined by the valence and number of the ions having octahedral coordination. A convenient method for calculating a formula is to reduce the analytical values of the ions having octahedral and tetrahedral coordination to their molal (M) values. It then is assumed that Al is distributed between tetrahedral and octahedral coordination so as to have $\Sigma = 2.00$. Thus $2(M(\text{Al})+M(\text{Fe}''')+M(\text{Fe}'')+M(\text{Mg})$ etc. $-y) = (M(\text{Si})+y)$ from which y, the molal amount of Al in tetrahedral coordination, can be evaluated. Amounts of the various ions in the formula then are obtained by multiplying their molal values by $4.00/(M(\text{Si})+y)$. The subscript (A) of X, the interlayer cation, is then fixed by the amount of charge required to balance the lattice. In general it will not be equal to the equivalence of the interlayer ions. It is brought into agreement by change of Al between octahedral and tetrahedral coordination, Σ being finally greater or less than 2.00. In only 2 of the 42 analyses used in this study is the value of Σ below 2 and in most of them it is slightly, but significantly above 2.

Glauconite specimen number 14 is treated here as an example:

	%	Divide by	Mols			Equivalents		
SiO_2	49.4	60.06	Si	.822				
Al_2O_3	10.2	50.99	Al	.200	Sum .555			
Fe_2O_3	18.0	79.84	Fe'''	.225				
FeO	3.1	71.84	Fe''	.043				
MgO	3.5	40.32	Mg	.087				
CaO	0.6	28.04				Ca	.021	Sum .174
K_2O	5.1	47.10				K	.108	
Na_2O	1.4	31.00				Na	.045	

The next step is to calculate y from the condition $2[.555-y] = [.822+y]$, which gives y equal to .096. Thus $.822+y = .916$, and the factor by which the molal quantities are to be multiplied to derive the trial formula is $4.00/.916 = 4.367$. This gives $.822\times4.367 = 3.59$ silicon atoms and a trial formula is $X_A[Al_{.46}Fe'''_{.98}Fe''_{.18}Mg_{.38}]$ $[Si_{3.59}Al_{0.41}]$ $0_{10}(OH)_2$. The sum of the cation valences indicated in the foregoing formula is 21.03, but the total cation valences must be 22 to balance the 10 oxygen ions, plus 2 hydroxyls. The difference of .97 must be supplied by Ca, K, and Na, and gives the value of X_A. However, the equivalence of Ca, K, and Na combined is only $.174\times4.367 = .76$. In order to bring these two quantities into agreement, aluminum must be transferred from tetrahedral to octahedral coordination. This is done by trial and error, always bearing in mind that as aluminum is transferred, the factor by which the molal amounts are to be multiplied changes. In this manner the formula is found to be:

$$X_{.75}[\mathrm{Al}_{.51}\mathrm{Fe}'''_{.99}\mathrm{Fe}''_{.18}\mathrm{Mg}_{.38}]\ [\mathrm{Si}_{3.63}\mathrm{Al}_{.37}]\mathrm{O}_{10}(\mathrm{OH})_2$$

Effect of Impurities

Submicroscopically crystalline micaceous minerals are very likely to contain fine-grained impurities. Appreciation of this factor, however, has led to an unnecessary assumption that impurities are so invariably present that no formula is justified.[23]

Glauconite being of sedimentary origin is commonly associated with coarse-grained detrital minerals, but that associated with clean crystalline limestones, or occurring in nearly pure beds, may be very free from accessory impurities. The glauconite can readily be concentrated by suspending it in water or by suitable heavy solutions, and only intergrown impurities need be feared. Possible fine-grained impurities are submicroscopically crystalline or amorphous phosphates and silica, leucoxene, calcium and iron carbonates, clay minerals, and oxides and hydrous oxides of iron. Small grains of phosphatic material (probably collophanite) are commonly associated with glauconite, and are only completely removed by careful hand picking. While many such impurities can now be recognized, much of the earlier work was carried out on more questionable material, and many of the modern glauconites associated with detrital materials are obviously quite impure. Clay minerals have a lower index of refraction and would be easily observable, and oxides and hydrous oxides of iron produce color changes warning of their presence. Sulfates, sulfides, chlorides, phosphates, or carbonates when present are adequate evidence of an impure sample.

Impurities that may be associated with celadonite are calcite, zeolites, saponite, serpentine, and cristobalite, common minerals in vesicular cavities of basalt, and oxides and hydrous oxides of iron resulting from subsequent oxidation. These impurities, with the exception of cristobalite and perhaps iron-rich saponite, are readily detected and probably were avoided in most samples selected for analysis. Cristobalite has to be accepted as a possible adventitious material in all samples which were not subjected to thorough microscopic examination.

Minor amounts of titanium and phosphorus have not been included in the calculations since it was thought that they were more likely to be present as impurities.

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[23] Galliher, E. W., Glauconite genesis: *Bull. Geol. Soc. Am.*, **46**, No. 9, 1351–1365 (1935).

CHEMICAL RELATIONS OF GLAUCONITE AND CELADONITE

Analyses of forty-one glauconite samples all taken from the literature, are listed in Table 2. These include fifteen samples previously considered by Ross[24] and nine samples analyzed by Glinka.[25] The remainder of the analyses were obtained from the sources indicated in the footnotes to the table. Eight recent analyses, made since 1930, are included among these.

Glinka carried out gravity separations using Thoulet's solution. This would induce replacement of exchangeable base,—which is of the order of one-tenth of the interlayer base,—by potassium, but should not otherwise be confusing if the heavy solution was completely removed. An excess of potassium would not effect the other constituents, and these are the ones that are most significant in the following calculations. Some of Glinka's samples probably contained calcium carbonate as an impurity, this being particularly true for samples 38 and 39 of Table 2. The small amount of calcium in samples 4 and 5 was also regarded as an impurity. Samples 13 and 32 from other sources also probably contained calcium carbonate as an impurity.

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[24] Ross, C. S., *loc. cit.*

[25] Glinka, K., *op. cit.*

TABLE 2. ANALYSES OF GLAUCONITE

	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)	(11)	(12)	(13)
SiO_2	47.88	47.6	48.5	51.00	49.53	49.0	50.70	49.12	46.70	49.75	48.66	49.09	49.67
Al_2O_3	14.94	9.9	9.0	9.93	5.84	9.2	19.80	7.09	5.52	7.82	8.46	18.19	9.29
Fe_2O_3	17.13	21.9	20.0	18.69	20.06	19.5	—	25.95	21.62	22.26	18.80	6.42	19.88
FeO	2.68	1.5	3.1	1.98	5.95	3.3	8.60	0.89	2.42	2.36	3.98	2.56	1.28
MgO	2.45	3.7	3.7	3.85	2.92	3.6	3.70	3.10	4.16	3.25	3.56	3.10	4.03
CaO	0.56	0.8	0.4	0.87	0.56	0.5	—	—	0.63	—	0.62	1.03	1.95
K_2O	8.04	5.3	6.1	7.66	9.31	6.3	8.20	7.02	6.97	9.01	8.31	5.62	3.68
Na_2O	0.43	1.4	1.5	0.35	0.46	0.9	0.50	—	1.73	0.30	0.00	0.23	3.00
H_2O (ign)	5.91	7.7	7.3	5.83	4.91	7.6	8.50	7.12	11.48	5.16	6.56	13.47	7.88
Total	100.02	99.8	99.6	100.16	99.54	99.9	100.00	100.29	101.23	99.91	99.08[a]	99.90[b]	100.66

[a] Including $P_2O_5=0.12\%$; $MnO=0.01\%$.

[b] Including $TiO_2=0.21\%$.

TABLE 2. (*Continued*)

	(14)	(15)	(16)	(17)	(18)	(19)	(20)	(21)	(22)	(23)	(24)	(25)	(26)
SiO_2	49.4	49.09	48.12	51.95	49.42	51.00	49.23	50.36	51.24	51.35	52.96	49.76	50.58
Al_2O_3	10.2	15.21	9.16	17.81	10.23	9.93	7.11	7.04	12.22	9.47	12.76	8.18	6.72
Fe_2O_3	18.0	10.56	19.10	6.17	16.01	18.69	20.89	19.13	13.44	16.37	13.56	16.00	19.50
FeO	3.1	3.06	3.47	3.87	3.00	1.98	3.06	3.95	3.06	4.75	2.34	3.77	2.96
MgO	3.5	2.65	2.36	4.76	3.78	3.85	3.44	4.08	3.93	3.17	4.11	3.97	4.10
CaO	0.6	0.55	0.76	0.53	0.31	0.87	tr	0.91	0.10	0.63	—	0.41	0.34
K_2O	5.1	6.05	7.08	4.04	7.91	7.66	8.51	6.62	7.50	7.34	8.69	7.57	8.26
Na_2O	1.4	1.21	0.22	1.06	0.26	0.35	0.11	1.58	0.31	1.22	0.47	0.52	0.04
H_2O (ign)	8.3	11.64	10.06	9.43	8.08	5.83	6.71	6.32	8.20	4.85	4.91	9.82	7.76
Total	99.6	100.02	100.33	99.81[c]	99.80[d]	100.16	99.06	100.34[e]	100.00	99.50[f]	99.80	100.00	100.83[g]

[c] Including $P_2O_5=.02\%$; $TiO_2=.10\%$; $MnO=.07\%$.

[d] Including insoluble$=0.80\%$.

[e] Including $TiO_2=.02\%$; $P_2O_5=.26\%$; $MnO=.06\%$; $Li_2O=.01\%$.

[f] Including $P_2O_5=0.35\%$.

[g] Including $P_2O_5=0.27\%$; $CO_2=0.30\%$.

TABLE 2. (*Continued*)

	(27)	(28)	(29)	(30)	(31)	(32)	(33)	(34)	(35)	(36)	(37)	(38)	(39)	(40)
SiO_2	51.15	49.47	51.11	52.74	51.55	49.81	55.95	51.90	54.58	50.62	46.90	42.32	41.02	47.59
Al_2O_3	7.61	5.59	4.88	12.29	4.03	6.33	11.56	1.52	7.17	3.80	4.06	16.51	22.19	17.99
Fe_2O_3	18.83	19.46	22.67	9.35	22.17	13.24	9.99	27.98	18.27	21.03	27.09	18.91	18.49	13.95
FeO	2.78	3.36	3.20	6.30	3.54	5.81	2.02	1.26	2.86	6.02	3.60	2.80	2.06	3.70
MgO	4.54	3.96	3.52	4.05	3.86	4.50	6.77	4.67	2.95	0.57[m]	0.70	1.74	0.69	1.80
CaO	—	0.60	0.52	0.55	0.69	1.58	3.95	0.89	1.86	0.54[m]	0.20	2.20	1.96	1.22
K_2O	7.80	8.04	6.88	7.97	7.03	7.45	4.12	4.90	5.34	7.14	6.16	7.49	5.74	7.21
Na_2O	—	0.16	0.63	0.09	0.55	0.16	0.61	0.53	1.23	—	1.28	0.42	0.38	0.42
P_2O_5	—	—	—	—	—	—	0.18	0.11	—	—	—	—	—	—
H_2O (ign)	7.56	8.54	6.34	5.93	6.62	9.70	4.82	6.15	6.53	9.14	9.25	7.48	7.88	5.27
Total	100.27	100.80[h]	99.75	99.40[i]	100.04	99.89[j]	99.97[k]	99.91[l]	99.48	99.86	99.24	99.87	100.41	99.15

[h] Including $P_2O_5=1.06\%$; $CO_2=0.56\%$.

[i] Including $F=0.13\%$.

[j] Including $P_2O_5=0.33\%$; $CO_2=0.98\%$.

[k] Including $TiO_2=0.59\%$.

[l] Including $TiO_2=0.30\%$.

[m] Including carbonates.

NOTE TO TABLE 2

C. O. Hutton and F. T. Seelye in an article on "Composition and properties of some New Zealand glauconites, *Am. Mineral.*, **26,** 595–604 (1941), give seven glauconite analyses. Their results calculated after the manner described in this paper lead to the following formulas:

Hutton and Seelye sample number	Tetrahedral coordination Si	Octahedral coordination				Interlayer cations X	No. of ions in octahedral coordination	Fe'''/Fe''
		Al	Fe'''	Fe''	Mg			
1	3.64	0.60	0.89	0.09	0.49	.73	2.07	9.9
2	3.89	0.39	1.00	0.22	0.38	.74	1.99	4.5
3	3.83	0.11	1.27	0.20	0.44	.75	2.02	6.3
4	3.46	0.13	1.49	0.19	0.35	.60	2.16	7.8
5	3.75	0.45	1.02	0.20	0.37	.71	2.04	5.1
6	3.55	0.18	1.29	0.41	0.25	.72	2.13	3.1
7	3.66	0.22	1.26	0.24	0.32	.78	2.04	5.2

The analyses apparently were made on very well selected materials and the results are in harmony with the general treatment of this paper.

TABLE 2—REFERENCES

1. Jurassic sandstone of Tschernofskoje. Glinka #5.
2. F(7) From unconsolidated Pleistocene west of San Pedro, California. Schneider, Hyrum, *Jour. Geol.*, **35,** 289–310 (1927).
3. F(5) From Franconia formation (Cambrian) near Norwalk, Wisconsin. Thomas B. Brighton, analyst. Schneider, Hyrum, *op. cit.*
4. Chalky sandstone of Padi. Glinka #1.
5. Eocene sandstone of Urals. Glinka #3.
6. F(4) From Hornerstown marl, Woodstown, New Jersey. Schneider, Hyrum, *op. cit.*
7. Island Orleans, Quebec. Hunt, T. S., *Geol. Canada* (**1863**), p. 487. Ross #4.
8. Pacific Ocean off Panama. Caspari, W. A., *Proc. Roy. Soc. Edinburgh,* **30,** 364–373 (1910). Ross #17.
9. F(8) From limestone near Palermo. Comucci, P., *Rend. Acc. Lincei Roma,* **30,** 220–224 (1921).
10. Chalky sandstone from Nasonovo. Glinka #2.
11. St. Joseph Lead Co. Mines. G. V. Brown, analyst. Ross #11.
12. Sandstone near Skole, Poland. Smulikowski, K., *Arch. Min. Soc. Sci. Varsovie,* **12,** 145–180 (1936); *Mineral. Abst.*, **6,** 345 (1936) (F).
13. F(1) From the Kupsten. Johnsen, A., *Scharf. Phys. Ökon Ges Königsberg,* **1,** 51–60 (1908).
14. F(6) From dolomite at the base of the Tempealeau (Upper Cambrian), northwest of Norwalk, Wisconsin. Schneider, Hyrum, *op. cit.*
15. Ashgrove, Elgin, Scotland. Heddle, M. F., *Trans. Roy. Soc. Edinburgh,* **29,** 79 (1879). Ross #15.
16. Lewes, Sussex. Radley, E. G., *Mineral. Mag.*, **19,** 331 (1920–22). Ross #12.
17. South Tyrol. Hummel, K., *Chemie der Erde,* **6,** 468–551 (1931).

18. Suir River, Olonets, Russia. Kupffer, A., *Archiv. Naturk, Liv.-, Ehst.- u. Kurlands, Ser I, Min. Wiss. Dorpat.*, **5**, 123 (1870). Ross #9.
19. Tertiary sandstone of Traktemiroff. Glinka #4.
20. Big Goose Canyon, near Sheridan Big Horn Mts., Wyoming. G. Steiger, analyst. *U. S. Geol. Survey, Bull.* **591,** 340 (1915). Ross #10.
21. Monte Brion, Lake Garda, Italy. *Sitzungsb. Akad. München*, **26,** 545 (1896). Ross #7.
22. Karga-Oro, Ontika, Esthonia. Kupffer, A., *Archiv. Naturk., Liv.-, Ehst.-, u. Kurlands, Ser I, Min. Wiss. Dorpat.*, **5,** 123 (1870). Ross #8.
23. F(2) From Silurian limestone, Eriksöre, Öland Sahlbom, Naima, *Geol. Inst. Univ. Upsala, Bull.* **15,** 211–212 (1916).
24. Lower Silurian limestone of Udriass. Glinka #10.
25. Grodno, Poland. Kupffer, A., *Archiv. Naturk., Liv.-, Ehst.-, u. Kurlands, Ser I, Min. Wiss. Dorpat.*, **5,** 123 (1870). Ross #2.
26. Sewell, N. J. Mansfield, G. R., *Econ. Geol.*, **15,** 557 (1920). Ross #5.
27. Agulhas Bank, South Africa. Caspari, W. A., *Proc. Roy. Soc. Edinburgh,* **30,** 364–373 (1910). Ross #6.
28. Elmwood Road, N. J. Mansfield, G. R., *Econ. Geol.*, **15,** 557 (1920). Ross #3.
29. Average analysis of seven samples from Tertiary. Takahashi, J. I., *Recent Marine Sediments*, 503–512 (1939).
30. F(3) From Schoen. Johnsen, A., *op. cit.*
31. Upper Cretaceous from Hokkaido. Takahashi, J. I., *Recent Marine Sediments*, 503–512 (1939).
32. F(9) From Marine Cretaceous, Zurawska region. Kampioni-Zakrzewska, *Ach. Min. Tow. Nauk Warsaw*, **13,** 9–17 (1937).
33. Monterey bay, California. A. A. Hanks, analyst. See number 34.
34. Galliher, E. W., *Bull. Geol. Soc. Am.*, **46,** 1359 (1935).
35. Recent sediment from Aomori Bay. Takahashi, J. I., *Recent Marine Sediments*, 503–512 (1939).
36. Havre, France. Haushofer, K., *Jour. Prakt. Chemie*, **102,** 38 (1886). Ross #1.
37. Agulhas Bank, South Africa. von Gumbel, C. W., *Sitzungsb. Akad. Wiss. München*, **16,** 417–449 (1886). Ross #16.
38. Jurassic sandstone of Karowo. Glinka #6.
39. Another fraction of 38. Glinka #7.
40. Jurassic sandstone of Kosolopowo. Glinka #9.

Formulas calculated from thirty-two of the glauconite analyses are listed in Table 3. Satisfactory formulas were not obtained for the remaining eight samples, which are listed in Table 2, chiefly to show the effect of impurities. Samples 33, 35, 38, 39, and 40 contained unduly large amounts of calcium, indicating a poor selection of material. Samples 33 and 35 further contained more SiO_2 than any of the other samples. Samples 33 and 34 are recent marine sediments that were known to be mixed with large amounts of other materials.[26] However, if sample 34 is predominantly glauconite, the low amount of Al_2O_3 that it contains would lead to formulas differing considerably from the ones obtained for the first thirty-two samples. Samples 36 and 37 are two samples previously discussed by Ross.[27] Formulas for them are:

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[26] Galliher, E. W., Glauconite genesis: *Bull. Geol. Soc. Am.*, **46,** 1351–1366 (1935).

[27] Ross, C. S., *loc. cit.*

(36) $X_{.80}(Al_{.21}Fe'''_{1.26}Fe''_{.40}Mg_{.07})(Si_{3.85}Al_{.15})O_{10}(OH)_2$
(37) $X_{.85}(Al_{.03}Fe'''_{1.59}Fe''_{.24}Mg_{.08})(Si_{3.65}Al_{.35})O_{10}(OH)_2$

Their MgO contents are very much less than that of the remaining samples and the Al_2O_3 contents also are low. The value of Σ, the total number of ions having octahedral coordination, for each sample is 1.94. While this is not an impossible situation it is indicative of an unusual condition of formation and might be the result of later alteration.

TABLE 3. GLAUCONITE FORMULAS

Sample number	Inter-layer cations X	Octahedral coordination				Tetrahedral coordination Si	No. of ions in octahedral coordination	Fe'''/Fe''
		Al	Fe'''	Fe''	Mg			
1	.86	0.69	0.93	0.16	0.26	3.44	2.04	5.8
2	.81	0.36	1.21	0.09	0.40	3.50	2.06	13.4
3	.83	0.35	1.12	0.19	0.40	3.58	2.06	5.9
4	1.00	0.25	1.28	0.08	0.33	3.58	1.94	16.0
5	1.00	0.14	1.17	0.39	0.33	3.61	2.03	3.0
6	.79	0.40	1.08	0.20	0.40	3.61	2.08	5.4
7	.78	1.27	0.51		0.39	3.61	2.17	
8	.66	0.22	1.43	0.05	0.34	3.61	2.04	28.6
9	.98	0.12	1.26	0.15	0.48	3.62	2.01	8.4
10	.87	0.29	1.22	0.14	0.35	3.62	2.00	8.7
11	.86	0.36	1.06	0.24	0.39	3.62	2.05	4.5
12	.72	1.20	0.35	0.16	0.34	3.63	2.05	2.2
13	.87	0.42	1.08	0.08	0.42	3.63	2.00	13.5
14	.75	0.51	0.99	0.18	0.38	3.63	2.06	5.5
15	.83	0.94	0.58	0.19	0.29	3.63	2.00	3.0
16	.83	0.44	1.08	0.22	0.27	3.63	2.01	4.9
17	.56	1.12	0.33	0.22	0.50	3.65	2.17	1.5
18	.83	0.54	0.90	0.19	0.41	3.65	2.04	4.7
19	.88	0.52	1.01	0.12	0.31	3.67	1.96	8.4
20	.81	0.28	1.18	0.19	0.38	3.67	2.03	6.2
21	.98	0.28	1.05	0.24	0.44	3.67	2.01	4.5
22	.73	0.73	0.73	0.18	0.42	3.69	2.06	4.1
23	.93	0.50	0.88	0.28	0.39	3.69	2.05	3.1
24	.84	0.76	0.71	0.14	0.43	3.71	2.04	5.1
25	.87	0.44	0.91	0.24	0.44	3.72	2.03	3.8
26	.85	0.31	1.08	0.18	0.45	3.72	2.02	6.0
27	.74	0.37	1.03	0.17	0.50	3.72	2.07	6.0
28	.91	0.24	1.11	0.21	0.44	3.74	2.00	5.3
29	.83	0.17	1.25	0.20	0.38	3.75	2.00	6.2
30	.78	0.79	0.50	0.36	0.44	3.75	2.09	1.4
31	.85	0.35	1.22	0.21	0.42	3.78	2.10	5.8
32	.86	0.41	*0.77*	*0.37*	*0.52*	*3.84*	2.07	2.1

TABLE 4. ANALYSES OF CELADONITE

	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)
SiO_2	55.61	53.23	50.6	54.30	54.73	52.69	55.30	50.70	57.72	56.41
Al_2O_3	0.79	2.13	4.2	5.08	7.56	5.79	10.90	4.72	0.33	2.14
Fe_2O_3	17.19	20.46	14.1	14.77	13.44	9.75	6.95	15.34	17.05	14.07
FeO	4.02	4.14	3.3	4.82	5.30	5.37	3.54	2.00	3.73	5.10
MgO	7.26	5.67	6.4	6.05	5.76	8.54	6.56	9.32	3.84	5.91
MnO	0.09	—	—	0.09	—	0.31	—	tr	0.08	0.23
CaO	0.21	—	3.1	0.80	0.00	1.16	0.47	1.32	0.60	0.60
K_2O	10.03	7.95	8.7	4.85	7.40	6.21	9.38	4.44	5.55	8.83
Na_2O	0.19	—	0.4	3.82	—	0.39	0.00	0.29	0.42	—
H_2O (ign)	4.88	6.18	8.3	5.64	6.40	10.48	6.51	12.52	10.78	6.80
Total	100.27	99.76	99.1	100.22	100.59	100.69	99.61	100.65	100.10	100.09

TABLE 4—REFERENCES

1. From vesicular basalt, 23 miles E. of Reno, Nevada. Wells, R. C., analyst. *U. S. Geol. Survey, Bull.* **878,** 102 (1937).
2. Bentonico, Monte Baldox, Italy. Levi, M. G., *Rivista di Min. e. Crist. Italiana,* **43,** 74 (1914).
3. From basalt, Sandoval Co., New Mexico. Wells, R. C., analyst. *U. S. Geol. Survey, Bull.* **878,** 102 (1937).
4. Madagascar. Raoult, M., analyst. Lacroix, M. A., *Soc. franc. Mineral. Bull.* **39,** 90–95 (1916).
5. Four miles E. of Vaile, Arizona. Koenig, G. A., *Jour. Phila. Acad. Nat. Sciences,* **15,** 424–425 (1912).
6. Amygdules in porphyritic basalt. Tayport Fife. Heddle, M. F., *Trans. Roy. Soc. Scot.,* **29,** 101–104 (1879).
7. From Vesuvius. Maegdefrau, E., and Hofmann, U., *Zeits. Krist.,* **A98,** 31–59 (1937).
8. Amygdaloidal basalt, Toselli pass Eriteria. Scherillo, A., *Periodico Min. Roma,* **9,** 253–264 (1938).
9. Cavities in basalt, Scuir Mohr Island of Rum. Heddle, M. F., *loc. cit.*
10. Druses in basalt S. of Grants Causway, Ireland. Heddle, M. F., *loc. cit.*

Analyses of ten celadonite samples, two of them new and the others taken from the literature, are listed in Table 4. Samples 9 and 10, which were studied by Heddle[28] probably contain a little silica as an impurity, and associated saponite was reported in No. 6. Calcium present in sample 4 was assumed to be present as the carbonate although it is a minor constituent. Sample 8 also contains an excess of magnesia.

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[28] Heddle, M. F., The minerals of Scotland: Celadonite: *Trans. Royal Soc. Scotland,* **29,** 102–104 (1879).

TABLE 5. CELADONITE FORMULAS

Sample number	Interlayer cations X	Octahedral coordination				Tetrahedral coordination Si	No. of ions in octahedral coordination	Fe'''/Fe''
		Al	Fe'''	Fe''	Mg			
1	.98	0.07	0.93	0.24	0.77	4.00	2.01	3.9
2	.72	0.08	1.13	0.25	0.62	3.91	2.08	4.5
3	.90	0.28	0.82	0.21	0.73	3.90	2.04	3.9
4	.96	0.30	0.80	0.29	0.64	3.88	2.03	2.7
5	.68	0.50	0.72	0.30	0.61	3.88	2.13	2.4
6	.78	0.37	0.55	0.33	0.95	3.88	2.22	1.7
7	.91	0.78	0.36	0.21	0.69	3.87	2.04	1.7
8	.67	0.20	0.87	0.12	1.05	3.78	2.24	7.3
Specimens definitely containing silica as an impurity. Maximum and minimum formulas are given								
9	.80	0.03	1.17	0.28	0.52	4.00	2.00	4.2
	.83	0.00	1.79	0.29	0.53	3.97	2.01	
10	1.00	0.19	0.81	0.33	0.67	4.00	2.00	2.5
	1.00	0.06	0.91	0.36	0.76	3.85	2.09	

GENESIS OF GLAUCONITE

The characteristics of glauconite occurrences are admirably described in the comprehensive works of Cayeux,[29] Collet and Lee,[30,31] and Hadding.[32]

Hadding from his critical review of the literature and his extensive observations on the glauconites of Sweden concluded: "On summing up the inferences that can be drawn from the investigation . . . we can say that glauconite is always marine, always sublittoral, always a shallow sea formation, as a rule formed in agitated water, as a rule formed under decreased deposition of detritus, often formed during negative sedimentation, . . . never formed in highly oxygenous water."

Ichimura[33] concluded from his examination of the glauconitic rocks of Formosa that the glauconite in general was formed from mud grains and is associated with the usual detrital minerals. He states: "Moreover, the

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[29] Cayeux, L., *op. cit.*

[30] Collet, L. W., and Lee, G. W., Recherches sur la Glauconi: *Proc. Roy. Soc. Edinburg.* **26,** 238–278 (1906).

[31] Collet, L. W., *Les Dêpots Marins,* 132–194, Paris (**1908**).

[32] Hadding, A., *op. cit.*

[33] Ichimura, T., *op. cit.*

mode of occurrence of glauconite grains suggests that they were subjected to sorting in some agitated water."

Twenhofel[34] concludes his review of the literature on glauconite with the statement: "The existing state of knowledge with respect to the origin of glauconite supports the view that it is a product of diagenesis and that the glauconitic particles were originally pellets of mud containing finely divided and colloidal clay and iron oxides; that in some as yet unknown manner the aluminum of the clay was removed and its place taken by colloidal iron, and potash, and colloidal silica was absorbed from sea water or surrounding materials . . . An environment intermediate between strongly reducing and strongly oxidizing seems necessary . . . "

There is no essential disagreement with the observation that glauconite forms only in a marine environment with muds as the primary material for its formation. It often is associated with decaying organic matter and pyrite is not infrequently present. Cayeux[35] noted three general types of occurrences: (1) associated with organic matter such as foraminifera, (2) pellets, and (3) pigmentary. Cayeux believed that organic matter was absent from the pellets and took this as evidence against necessary presence of organic matter in the formation of glauconite. Buchanan[36] and Takahasi and Yagi,[37] however, noted various degrees of glauconitization of coprolites and it is the current opinion that some glauconite pellets were coprolites. Hadding presents evidence that the glauconite grains were initially colloidal material that had been transported; some being still colloidal when incorporated into the sedimentary beds, while others had undergone crystallization.

Galliher[38] recently has noted the associtaion of glauconite with partially altered biotite and has suggested that green sand deposits formed from biotite. Murray and Phillippi[39] had earlier suggested that glauconite was derived from micas and potassium feldspars. Derivation from biotite can at most have only local application, and is out of harmony with earlier observations. Several objections to such an origin in general may be pointed out.

Mineral associations in many green sands indicate that large supplies

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[34] Twenhofel, W. H., *Treatise on Sedimentation,* Baltimore, 453–460 (**1932**).

[35] Cayeux, L., *op. cit.*

[36] Buchanan, J. Y., On the occurrence of sulphur in marine muds, and nodules and its bearing on their mode of formation: *Proc. Roy. Soc. Edinburgh,* **18,** 19–20 (1890).

[37] Takahasi, J., and Yagi, T., The peculiar mud-grains in the recent littoral and estuarine deposits, with special reference of the origin of glauconite: *Econ. Geol.*, **24,** 838–852 (1929).

[38] Galliher, E. W., *loc. cit.*

[39] Murray, J., and Phillippi, E., *Die Grundprolien der deutschen Tiefsee-Exped. auf dem Dampfer "Valdiva," 1898–1899,* **10,** 177 (1908).

of micas and feldspars are not necessary for glauconite formation.[40] Some of the green sand beds of New Jersey are many feet in thickness and are composed of nearly pure glauconite. The accumulation of biotite in such thickness and freedom from impurities would be impossible. The almost complete absence of normal detrital materials and especially of muscovite, the dominant mica in all materials derived from crystalline rocks, and a material resistant to alteration under marine conditions, is especially significant.

Glauconite has formed in association with biotite in arkosic Ordovician bentonites in Tennessee; that is, a bentonite whose characteristic mineral is a hydrous mica. The biotite which is nearly fresh in most Ordovician bentonite is bleached in this occurrence. It has acted as a locus for the deposition of glauconite so that the two minerals are in parallel orientation, but there has been no replacement of biotite by glauconite.

A recent study of rocks from the Panama Canal Zone composed of andesitic rock debris, has shown the presence of normal glauconite in rounded grains, together with another type that has formed by direct replacement of andesite rock grains and of augite.

Ross, Miser, and Stephenson[41] mention glauconite in association with volcanic materials from southwestern Arkansas. This material has formed under marine conditions and so its genesis is that of marine glauconite. This material has most commonly formed as a replacement of grains of phonolitic rock as shown by the mottled grains in the upper part of Plate 1, Fig. *a*; the gray portion being glauconite. Less commonly, glauconite has replaced augite as in the large grain at the bottom of the figure. The light gray interstitial material is calcite. In a few specimens glauconite has filled spaces between mineral and rock grains, where it has a structure indicating crystallization from material originally deposited as a colloid. In none of these occurrences is there any paragenetic relation to biotite. The origin of glauconite has also been discussed by Hadding.[42]

Attention is now directed to the glauconite formulas of Table 3, which were derived from analyses of specimens from many different deposits. They nevertheless show some remarkable regularities indicative of the common mode of origin for sedimentary glauconites. The magnesium content, referred to the general formula, is between 0.35 and 0.45 ion for twenty of the thirty-two analyses, and the maximum variation is

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[40] Note also, Correns, C. W., *Die Sedimentgesteine*, p. 209, Berlin (**1937**).

[41] Ross, Clarence S., Miser, Hugh D., and Stephenson, Lloyd D., Waterlaid volcanic rocks of early Upper Cretaceous age in southwestern Arkansas, southeastern Oklahoma, and northwestern Texas: *U. S. Geol. Surv.*, *Prof. Paper* **154,** 184 (1928).

[42] Hadding, A., *op. cit.*

from 0.26 to 0.52 ion. Silicon in tetrahedral coordination varies from 3.44 to 3.84 ions in the thirty-two samples, and all except four samples have between 3.58 and 3.75 ions. The interlayer ion content is definitely less than one equivalent, the mean value being 0.84. An average formula is:

$$(K,Ca_{1/2},Na)_{.84}(Al_{.47}Fe'''_{.97}Fe''_{.19}Mg_{.40})(Si_{3.65}Al_{.35})O_{10}(OH)_2$$

Muds in general and all the finely divided detrital minerals, with the exception of hydrous micas, contain less magnesium than required by the above formula. An average formula derived from six hydrous mica analyses listed by Grim, Bray, and Bradley[43] is:

$$K_{.58}(Al_{1.38}Fe'''_{.37}Fe''_{.04}Mg_{.34})(Si_{3.41}Al_{.59})O_{10}(OH)_2$$

The hydrous micas vary in composition, the limits of which are undetermined, but the foregoing formula is probably representative enough to show the similarities and striking differences from glauconite.

The improbability of derivation from a material carrying adequate potassium, and the invariable formation under a marine environment, indicates that potassium was derived from the sea, and the same is, no doubt, true of magnesium. In this connection it may be pointed out that a bentonite from Mine Creek, Howard County, Arkansas, with several per cent of MgO, has formed from volcanic ash that contained but a fraction of a per cent of MgO; and since alteration occurred in a marine embayment, sea water was no doubt the source of the excess magnesium.[44]

The near constancy in the magnesia content of glauconites to 0.32 reflects the essentially unchanging nature of the environment and the structural requirements of the mineral lattice. The other constituents of glauconite, silicon, aluminum, and iron could adequately be supplied by any mud. There is thus no particular restriction on the type and composition of the source material.

The amount of aluminum having octahedral coordintaion in glauconite varies from 0.12 to 1.27 ions, with accompanying variations in the total amount of iron. It clearly is not constant and probably is not determinative in the formation of the mineral. Ferrous iron is characteristic of glauconite, and the value of Fe'''/Fe'' for half the samples is between 4.1 and 6.2. These are narrow limits considering the factors involved and the possibility of subsequent oxidation. However, it probably is not due to a particular reducing condition but rather to several different factors. Among these would be the requirements of the structure; and the possible

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[43] Grim, R. E., Bray, R. H., and Bradley, W. F., Mica in argillaceous sediments: *Am. Mineral.*, **22,** 813–829 (1937).

[44] Ross, Clarence S., Miser, Hugh D., and Stephenson, Lloyd D., *op. cit.*, p. 187.

precipitation of iron sulfide, which limits the concentration that ferrous iron could reach in the system in which glauconite is forming.

Factors influencing the availability of ferrous iron in a marine environment have been thoroughly considered by Cooper.[45] He shows that the potential in a system in which the equilibrium $Fe^{+++}+e^{-}\rightleftarrows Fe^{++}$ is established is given by $-E=[1.011-0.058 \log A_{Fe^{++}}-0.174\ pH]$ volts, where $A_{Fe^{++}}$ is the activity of the ferrous iron and *pH* is the usual negative log of the hydrogen ion concentration. The system is assumed to be saturated with respect to the ferric ion. If the potential is -0.4 volt (which is strongly reducing) and the *pH* is 6.0 (which is as acid as the marine environment reasonably could be) the ferrous ion activity would be 4×10^{-8} gm. mols. per liter, which is very dilute. The oxidation reduction potential for the reaction $SO_4^{--}+4H_2O\rightleftarrows S^{--}+4H_2O(l)$ calculated from the standard free energies of SO_4^{--}, S^{--}, and $H_2O(l)$ is -0.14 volt. This is a much stronger reducing system than the ferric, ferrous system and will be adequate at any reasonable hydrogen ion concentration to reduce the iron.

The not uncommon occurrence of ferrous sulfide minerals with glauconite indicates that sulfate reducing bacteria might often create the necessary oxidation reduction potential for glauconite formation. Bacteria commonly are found in muds[46] and could use organic materials of plant and animal remains and coprolite pellets as food. However, this is equally true of other bacteria that reduce ferric compounds to ferrous ones.[47] The only essential factor is strongly reducing conditions, including almost complete absence of oxygen, and this necessarily requires the action of bacteria on organic matter.

The important point of these considerations is that ferrous iron necessary for glauconite formation appeared only as a result of bacterial action in a system of negligible oxygen content, that is, anaerobic. The ferrous iron concentration in solution was very small and the ferrous iron was fixed in some insoluble form having much lower solubility than the hydroxide, possibly in glauconite itself. However, it is probable that before or during the complete formation of glauconite the portion of the system containing ferrous iron was sometimes transported as a colloid.

The number of ions having octahedral coordination, Σ, varies between 2.00 and 2.09 for all except five of the samples. Glauconite thus is a hep-

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[45] Cooper, L. H. N., Some conditions governing the solubility of iron: *Proc. Roy. Soc.* (London), B, **124,** 299–307 (1938).

[46] Bastin, E. S. (Collaboration of Anderson, B., Greer, F. E., Merritt, C. A., and Moulton, G.), The problem of the natural reduction of sulfates: *Bull. Am. Soc. Petroleum Geologists* **10,** 1270–1299 (1926).

[47] Buchanan, R. E., and Fulmer, E. I., *Physiology and biochemistry of bacteria,* Baltimore, **3,** 211–216 (1930).

taphyllite type of mica showing very limited solid solution toward an octaphyllite (biotite) type, despite the presence of Mg″ and Fe″ to the extent of $\frac{1}{4}\Sigma$ to $\frac{1}{3}\Sigma$. The average amount of aluminum in tetrahedral coordination is 0.35 ion which is greatly less than the amount, near 1.00 ion, found in the usual micas. The number of interlayer cations, K′, Ca″, Na′, is determined by these interrelations and while the maximum possible value is 1.00, it is not surprising to find actual amounts slightly less.

If glauconite is formed in the presence of normal ocean waters, then the ratios of the amounts of the interlayer ions are determined by competition, depending upon their affinity for the available sites in the crystal lattice and their concentration in the medium. The average interlayer ionic ratios in equivalents for glauconite are near

$$\mathrm{Na:Ca:K::0.15:0.20:1.0}$$

A few samples deviate considerably from these ratios, which is not unexpected since glauconite did not necessarily form in normal ocean water or might have undergone later exchange of alkalis, although as previously pointed out there is a strong indication of a nearly uniform genetic environment for most of the samples. Ionic ratios in equivalents in ocean water are[48] Na:Ca:K::46.3:2.1:1.0. These combined into an affinity series give about Na:Ca:K::300:10:1. In other words for glauconite to contain equal amounts of Na′ and K′ the solution in contact would have to contain 300 times as much Na′ as K′. The chemistry of the system from which micas form is not experimentally reproducible, and so glauconite formation under marine conditions where the chemistry of the system is approximately known is significant because it serves to illustrate the dominant formation of potash micas even from systems that were obviously rich in sodium.

Genesis of Celadonite

Formulas derived from analyses of celadonite are listed in Table 5. Samples 9 and 10 contained excess silica and for this reason maximum and minimum amounts of silicon in tetrahedral coordination were assumed. The maximum amount of course is 4.00 ions for the usual formula, and the minimum is that amount consistent with the amounts of the remaining constituents. These limits serve to emphasize the maximum possible effect of impurities.

There are three prominent features of these celadonite formulas. The magnesium content again is surprisingly constant, the average value per formula being 0.76 ion with three-fourths of the analyses in the range

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[48] Clarke, F. W., The Data of Geochemistry: *U. S. Geological Survey, Bull.* **770,** 127 (1924).

0.61–0.77 ion. In none of the samples is the Mg'' content as low as the highest value found for glauconite. Silicon in tetrahedral coordination is very high, the minimum value again being greater than the maximum value for glauconite. The total iron content varies greatly but a large fraction of it is ferrous which leads to a smaller value for Fe'''/Fe'' than generally found for glauconite. An average formula is:

$$(K,Ca_{1/2},Na)_{.82}(Al_{.34}Fe'''_{.76}Fe''_{.24}Mg_{.76})(Si_{3.89}Al_{.11})O_{10}(OH)_2$$

Celadonite is essentially a heptaphyllite mica, Σ, the number of ions in octahedral coordination, varying from 2.01 to 2.24, but it could not safely be held that Σ is truly greater than 2.13. Despite the heptaphyllite character about half the ions having octahedral coordination are doubly charged, Mg'' and Fe''. The lack of intralayer charge requiring presence of interlayer ions is almost entirely confined to positions with octahedral coordination. Samples number 1, the excellent specimen from the vicinity of Reno, Nevada, and number 9 are of particular interest in that they indicate that aluminum is not a necessary constituent of celadonite. Limiting formulas for samples number 1 and 9 could be:

$$(1)\ K_{1.0}(Fe'''_{1.00}Fe''_{.25}Mg_{.75})Si_{4.0}O_{10}(OH)_2$$
$$(9)\ K_{0.8}(Fe'''_{1.20}Fe''_{.30}Mg_{.50})Si_{4.0}O_{10}(OH)_2$$

Even though sample number 7 contains more aluminum than seven-tenths of the glauconite samples, it has less in tetrahedral coordination than any of them. This apparently is due to the higher magnesium content of celadonite which further emphasizes the determinative role of magnesium in the formation of glauconite.

A study of the rocks in which celadonite occurs gives significant information about the chemical relations which control its formation. The parent rock is usually a vesicular basalt, commonly an olivine basalt. Celadonite not only fills vesicular cavities in basalt but directly replaces olivine in a number of specimens. This is shown in Plate 1, Fig. *b*, where a phenocryst of olivine has been partly replaced by celadonite (gray material in upper portion), while the lower part represents only partly altered olivine. Olivine, $(Mg,Fe)_2SiO_4$, is an unstable mineral in the presence of water vapor and commonly is altered in the late cooling (deuteric) stages of a basalt. In most of the basalt specimens examined that contained celadonite, olivine has in general been largely altered to serpentine, a hydrous silicate of magnesium which often contains ferrous iron. On the other hand, iddingsite, another widely occurring deuteric alteration product of olivine, but one characterized by ferric iron,[49] is not

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[49] Ross, Clarence S., and Shannon, Earl V., The origin, occurrence, composition, and physical properties of the mineral iddingsite: No. 2579, From the *Proc. U. S. Nat. Museum*, **67**, 1–19 (1925).

associated with celadonite, apparently because of the oxidizing conditions during its formation.

Celadonite, as well as glauconite, in many specimens has the appearance of having passed through a gel state in its formation, that is, it is now a metacolloid. Olivine in altering to celadonite could supply silicon, magnesium, and ferrous iron. However, some magnesium would have to be removed. Part of the ferrous iron would have to be changed to ferric iron, but the remaining ferrous iron would require that the environment be a reducing one. It would be necessary for all the potassium, as well as the aluminum, to be introduced from nearby material. On the other hand, material forming celadonite in vesicular cavities would all have to be transported, at least for short distances.

END MEMBER FORMULAS FOR GLAUCONITE AND CELADONITE

It is desirable when possible to give end formulas for minerals of variable composition. Derivation of a reasonable number of suitable end formulas for glauconite and celadonite, however, is a simplification since the composition of each is influenced by at least six variables. These variables are Al, Fe''', Fe'', and Mg having octahedral coordination, the number of ions between layers, and the amount of Al having tetrahedral coordination. A rigorous representation of the composition takes account of the fact that glauconite and celadonite are not strictly heptaphyllitic (the ions in octahedral positions normally exceed 2) and the interlayer bases (essentially K) are in general less than 1. These two factors are neglected in the end members represented in the following list. However, the representation of the composition by such end members is sufficiently close to illustrate the relationships of glauconite and celadonite within the mica group.

The most unusual features of glauconite and celadonite as essentially heptaphyllite micas is their considerable content of ferric iron and the predominance of silicon in tetrahedral coordination. Compositions of these minerals can approximately be represented by combination of the following six formulas, listed in the order of their abundance:

(1) $K(Fe'''Mg)Si_4O_{10}(OH)_2$
(2) $K(Fe'''Fe'')Si_4O_{10}(OH)_2$
(3) $K(AlMg)Si_4O_{10}(OH)_2$
(4) $K(AlFe'')Si_4O_{10}(OH)_2$
(5) $K(Fe''')_2AlSi_3O_{10}(OH)_2$
(6) $K(Al)_2AlSi_3O_{10}(OH)_2$

From these six formulas all possible combinations of Al, Mg, Fe''', and Fe'' in strictly heptaphyllite micas can be obtained.

Formula number (6) is that usually given for muscovite and number

(5) represents a ferric analogue of muscovite.[50] Analyses of sericites seem to trend towards the composition represented by formula (3). This formula and the remaining three formulas (1), (2), and (4) are characteristic of celadonite. Thus celadonite, sample number (1), can be represented by .77(1)+.16(2)+.07(4), and sample number (5), approximately by .58(1)+.20(4)+.12(6)+.10(2). Table 6 shows the close agreement between the chemical analyses and the compositions (in weight percentage) calculated on the basis of the foregoing interpretation; celadonites 1 and 5 being arbitrarily selected as examples:

TABLE 6. INTERPRETATION OF COMPOSITION OF CELADONITES 1 AND 5

Percentage	Celadonite 1	Percentage	Celadonite 5
77	$KFe'''MgSi_4O_{10}(OH)_2$	58	$KFe'''MgSi_4O_{10}(OH)_2$
16	$KFe'''Fe''Si_4O_{10}(OH)_2$	10	$KFe'''Fe''Si_4O_{10}(OH)_2$
7	$KAlFe''Si_4O_{10}(OH)_2$	19	$KAlFe''Si_4O_{10}(OH)_2$
—		13	$KAl_2AlSi_3O_{10}(OH)_2$
100	Total	—	
		100	Total

	Celadonite 1 Analysis	Celadonite 1 Calculated	Celadonite 5 Analysis	Celadonite 5 Calculated
SiO_2	55.61	55.82	54.73	54.55
Al_2O_3	0.79	0.83	7.56	7.25
Fe_2O_3	17.19	17.24	13.44	12.63
FeO	4.02	3.69	5.30	4.76
MnO	0.09		—	—
MgO	7.26	7.30	5.76	5.50
CaO	0.21		0.00	—
K_2O	10.03	10.94	7.40	11.08
Na_2O	0.19		—	—
H_2O	4.88	4.18	6.40	4.23
	100.27	100.00	100.59	100.00

That the celadonite type of formula, namely: $K(Fe''', Al)(Mg, Fe'')Si_4O_{10}(OH)_2$ greatly predominates in all the celadonites, is shown in Table 7. Analyses 6 and 8 must have been made on impure material, as a magnesium silicate (serpentine or saponite?) must have been present to account for the high percentages of MgO. These two analyses hence are omitted.

The last two columns show the great preponderance of the celadonite type of formula over that of the muscovite type, in all celadonites.

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[50] Clarke, F. W., and Darton, N. H., On a hydromica from New Jersey: *U. S. Geol. Surv., Bull.* **167,** 155 (1880).

TABLE 7. INTERPRETATION OF ANALYSES OF CELADONITE

No.	Celadonite $K(Fe''', Al)(Mg, Fe'')Si_4O_{10}(OH)_2$ type				Muscovite $K(Fe''', Al)_2AlSi_3O_{10}(OH)_2$ type		Cela- donite type	Musco- vite type
	Fe'''Mg	Fe'''Fe''	AlMg	AlFe''	Fe'''	Al		
1	77	16	—	7	—	—	100	0
2	60	26	—	—	13	1	86	14
3	70	9	—	13	—	8	92	8
4	62	16	—	13	—	9	91	9
5	58	10	—	20	—	12	88	12
7	37	—	30	21	—	12	88	12
9	56	31	—	—	13	—	87	13
10	66	15	—	19	—	—	100	0

In the interpretation of the composition of the glauconites on this basis, more than one distribution of Al and Fe''' as between formulas 5 and 6 is possible. Glauconite 1 is arbitrarily taken as an example. All of the alumina may be considered as belonging to the muscovite type of formula, $K(Fe''', Al)_2AlSi_3O_{10}(OH)_2$, as given in Table 8, column 1, when the remaining constituents belong to the celadonite type of formula, $KFe'''(Mg, Fe'')Si_4O_{10}(OH)_2$ free from alumina. In the second interpretation the alumina is divided between the two types of formulas, as shown in column 3.

TABLE 8. INTERPRETATION OF GLAUCONITE 1

Per- centage	1 Calculated	2	Per- centage	3 Calculated
26	$KFe'''MgSi_4O_{10}(OH)_2$	Analysis	24	$KAlMgSi_4O_{10}(OH)_2$
17	$KFe'''Fe''Si_4O_{10}(OH)_2$		16	$KAlFe''Si_4O_{10}(OH)_2$
27	$KFe'''_2AlSi_3O_{10}(OH)_2$		49	$KFe'''_2AlSi_3O_{10}(OH)_2$
30	$KAl_2AlSi_3O_{10}(OH)_2$		11	$KAl_2AlSi_3O_{10}(OH)_2$
100	Total		100	Total
SiO_2	47.87	47.88		47.86
Al_2O_3	14.54	14.94		14.69
Fe_2O_3	17.30	17.13		17.15
FeO	2.67	2.68		2.69
MgO	2.47	2.45		2.44
CaO	—	0.56		—
K_2O	10.96	8.04		10.97
Na_2O	—	0.43		—
H_2O	4.19	5.91		4.20
	100.00	100.02		100.00

In either interpretation the muscovite type of formula which played only a minor role in the composition of the celadonites, now becomes of equal importance with the celadonite type of formula and in the interpretation shown in column 3, the ferric iron analogue of muscovite is the dominant component.

An arbitrarily selected group of glauconites, so chosen as to include a wide range in composition, may be approximately interpreted as having the following component composition:

TABLE 9. INTERPRETATION OF SELECTED ANALYSES OF GLAUCONITE

No.	Celadonite $\mathrm{K(Fe''', Al)(Mg, Fe'')Si_4O_{10}(OH)_2}$ type				Muscovite $\mathrm{K(Fe''', Al)_2AlSi_3O_{10}(OH)_2}$ type		Cela-donite type	Musco-vite type
	Fe'''Mg	Fe'''Fe''	AlMg	AlFe''	Fe'''	Al		
1	26	17	—	—	27	30	43	57
5	31	38	—	—	22	9	69	31
8	37	—	—	7	56	—	44	56
10	36	15	—	—	39	10	51	49
12	36	—	—	17	—	47	53	47
21	44	—	—	24	30	2	68	32
31	41	23	—	—	36	—	64	36

The marked increase of importance of the muscovite type of formula in the glauconites is shown in the last two columns.

This study has confirmed the close relationship of glauconite and celadonite and raises the question of retention of both names. While the composition of neither of these members of the mica group can be expressed by one formula, the essentially aluminum-free celadonites are dominantly $\mathrm{KFe'''(Mg, Fe'')Si_4O_{10}(OH)_2}$ with Mg greatly in excess over Fe''. Glauconites, on the other hand, do not approximate any single simple formula but represent a much more extended series whose characteristics seem to be an approximate composition of about equal parts of the celadonite type formula, as given above, and the muscovite type of formula, with the ferric analogue of muscovite predominating.

As the well established term "glauconite" is used for a mineral of characteristic sedimentary origin whereas the term "celadonite" is used for a mineral of quite different occurrence and paragenesis, it seems best to retain both names.

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