

## PHILLIPSITE FROM SILICIC TUFFS IN SALINE, ALKALINE-LAKE DEPOSITS

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**Abstract**—Phillipsite is a common constituent in diagenetically altered rhyolitic vitric tuffs of Cenozoic saline, alkaline-lake deposits where it occurs as spherulites or aggregates of prismatic crystals. This phillipsite has chemical and physical properties that differ from phillipsites that occur in rocks of other compositions and depositional environments, but previously it had not been completely characterized. Published analyses of five samples from four deposits in the southwestern United States show that the phillipsite is very siliceous and alkalic; the Si/(Al + Fe<sup>3+</sup>) ratio is 3.08–3.37, alkalis greatly exceed the divalent exchangeable cations, and the Na/K ratio is 1.05–3.05. The mean index of refraction (1.451–1.470) is much lower than that generally reported for the phillipsite group and can be correlated with the relatively high Si and alkali contents. Monoclinic unit-cell parameters show the following ranges:  $a = 9.931\text{--}9.991$  Å,  $b = 14.142\text{--}14.155$  Å,  $c = 8.650\text{--}8.706$  Å,  $\beta = 124.57\text{--}125.07^\circ$ , and  $V = 1000.3\text{--}1007.4$  Å<sup>3</sup>. The especially small  $b$  dimension is consistent with the high Si content.

**Key Words**—Alkaline lake deposit, Chemical composition, Phillipsite, Tuff, Unit-cell parameters, X-ray powder diffraction, Zeolite.

### INTRODUCTION

Zeolites occur in rocks that are diverse in lithology and age and have formed in many different geological environments. Zeolites are among the most common authigenic silicate minerals reported in sedimentary deposits, especially those that originally contained volcanic glass (Hay, 1966). Phillipsite is one of about 20 different zeolites now recognized as occurring in sedimentary deposits. The common occurrences of diagenetic phillipsite in deposits of saline, alkaline lakes was documented by Hay (1964), more than 70 years after phillipsite was first discovered in sedimentary deposits by Murray and Renard (1891) in deep-sea sediments from the Pacific Ocean.

Hay (1964) described the chemistry, optical properties, and mineralogical associations of phillipsite in rhyolitic vitric tuffs from several saline, alkaline-lake deposits. Among other observations, he recognized that these phillipsites are the most siliceous ones ever reported and that they have indices of refraction considerably lower than those previously determined for phillipsites of other origins. Although phillipsite in rhyolitic vitric tuffs of saline, alkaline-lake deposits locally forms nearly monomineralic beds, it commonly coexists with other diagenetic silicate minerals, including other zeolites, smectite, potassium feldspar, searlesite, opal-CT, and quartz. Phillipsite has been reported to coexist with analcime, chabazite, clinoptilolite, erionite, and mordenite, but the association with clinoptilolite is most common (Sheppard and Gude, 1968, 1969). Where associated with other diagenetic zeolites, phillipsite was generally the earliest zeolite to crystallize. The phillipsite and coexisting zeolites crystallized from pore fluids that originated from the dissolution of rhyolitic glass by the saline, alkaline lake water (Hay, 1964).

The present study was undertaken to provide better characterization of phillipsite from the silicic, vitric tuffs. New indices of refraction and cell parameters were determined, and a special effort was made to correlate the chemistry with the unit-cell parameters. These highly siliceous phillipsites from silicic, vitric tuffs of saline, alkaline-lake deposits were the only group of phillipsites for which unit-cell parameters were lacking (Gottardi and Galli, 1985, p. 140).

### MATERIALS AND METHODS

The phillipsites used in the present study occur in diagenetically altered, rhyolitic vitric tuffs of Miocene to Pleistocene saline, alkaline-lake deposits. Splits of five samples, which had been previously chemically analyzed, were obtained for the additional studies. The chemical analyses had been performed by classical whole-rock techniques (Peck, 1964) on pure or nearly pure phillipsite separates (Table 1). Optical examinations of the phillipsites were made using immersion oil mounts. All measurements of the indices of refraction are considered accurate to  $\pm 0.001$ . A Cambridge Stereoscan 250 MK2 scanning electron microscope was used to study freshly broken fragments of the phillipsite-rich tuffs, which had been coated with a film of gold (about 300 Å thick) just prior to examination.

X-ray powder diffraction data (CuK $\alpha$  radiation) on ground (<45  $\mu\text{m}$ ) samples were collected using a Siemens D-500 automated diffractometer. Step scans were made over  $5^\circ\text{--}70^\circ 2\theta$  using a step size of  $0.02^\circ 2\theta$  and a 1-s integration time. The data were background stripped, smoothed, and peak searched using the modified Lorentzian simplex fit of Mallory and Snyder (1979). Miller indices were assigned to reflections using two approaches. First, the data were scanned for peaks

Table 1. Chemical analyses and unit-cell contents of phillipsite from rhyolitic vitric tuffs of saline, alkaline-lake deposits.

	1	2	3	4	5
Chemical analyses (wt. %)					
SiO <sub>2</sub>	56.55	57.48	56.84	58.95	56.60
Al <sub>2</sub> O <sub>3</sub>	13.23	14.29	14.82	15.14	15.35
Fe <sub>2</sub> O <sub>3</sub>	1.67	0.78	0.52	0.92	0.42
FeO	0.07	0.00	0.00	0.11	0.00
MgO	2.47	0.22	0.10	0.62	0.27
CaO	0.48	0.01	0.00	1.37	0.07
BaO	0.35	—	—	—	—
SrO	0.39	—	—	—	—
Na <sub>2</sub> O	4.14	4.51	5.42	3.72	6.95
K <sub>2</sub> O	5.69	6.55	5.81	5.18	3.48
H <sub>2</sub> O+	6.83	8.67	8.92	6.68	9.64
H <sub>2</sub> O-	6.74	7.00	7.01	7.24	6.90
TiO <sub>2</sub>	0.20	0.10	0.13	0.07	0.26
P <sub>2</sub> O <sub>5</sub>	0.19	0.00	0.01	—	0.01
MnO	0.02	0.00	0.01	—	0.00
Total	99.02	99.61	99.59	100.00	99.95

Unit-cell contents based on 32 oxygen atoms  
(Fe<sup>2+</sup>, Ti, P, and Mn were omitted in calculation  
of the unit cell)

	1	2	3	4	5
Si	12.02	12.26	12.16	12.17	12.06
Al	3.31	3.59	3.74	3.68	3.85
Fe <sup>3+</sup>	0.26	0.13	0.08	0.15	0.07
Mg	0.78	0.07	0.03	0.19	0.09
Ca	0.11	0.00	0.00	0.30	0.02
Ba	0.03	—	—	—	—
Sr	0.05	—	—	—	—
Na	1.70	1.87	2.25	1.49	2.87
K	1.54	1.78	1.59	1.36	0.94
H <sub>2</sub> O	9.62	11.15	11.36	9.58	11.75
Si + Al + Fe <sup>3+</sup>	15.59	15.98	15.98	16.00	15.98
Si : (Al + Fe <sup>3+</sup> )	3.37	3.30	3.18	3.18	3.08
Si : (Si + Al + Fe <sup>3+</sup> )	0.77	0.77	0.76	0.76	0.75

1. Miocene to Pliocene Big Sandy Formation, near Wikieup, Mohave County Arizona; field no. W7-25A (Sheppard and Gude, 1973, p. 19).

2. Pleistocene Lake Tecopa, near Shoshone, Inyo County, California; field no. T5-1-2 (Sheppard and Gude, 1968, p. 15).

3. Pleistocene Lake Tecopa, near Tecopa, Inyo County, California; field no. T4-49A (Sheppard and Gude, 1968, p. 15).

4. Upper Cenozoic Waucoba Beds of Hopper (1947), near Big Pine, Inyo County, California; field no. 480-80 (Hay, 1964). Analysis corrected for minor quartz, feldspar, and calcite impurities and recalculated to 100%.

5. Miocene Barstow Formation, Mud Hills, San Bernardino County, California; field no. M4-5A (Sheppard and Gude, 1969, p. 15).

which could be unambiguously indexed. A first trial for approximate unit-cell parameters was then calculated with the least-squares algorithm of Appleman and Evans (1973), utilizing these assigned reflections and a wide tolerance (0.08°2θ) for acceptance of unindexed reflections. After this pass, several additional unambiguously reflections generally could be identified. These reflections were then fixed for a second pass with a

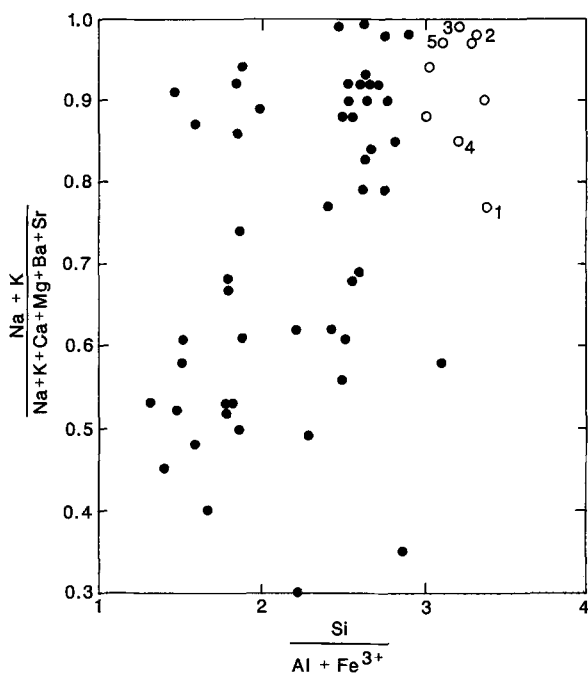


Figure 1. Plot showing the compositional range of phillipsite samples. Open circle, phillipsite from rhyolitic tuffs of saline, alkaline-lake deposits; solid circle, phillipsite from a variety of other sedimentary and igneous rocks. Numbered open circles are analyses listed in Table 1. Sources for the other analyses are Hay (1964, 1966), Regis and Sand (1966), Sheppard *et al.* (1970), Galli and Loschi Ghittoni (1972), Stonecipher (1978), and Passaglia and Vezzalini (1985).

narrower acceptance window (0.04°2θ). After all unambiguously indexed reflections were located, those having multiple indexing were resolved whenever possible using intensities calculated from the monoclinic structure of Rinaldi *et al.* (1974). This calculation was made using the chemistry and unit-cell parameters for our material. Multiple indexing for some peaks could

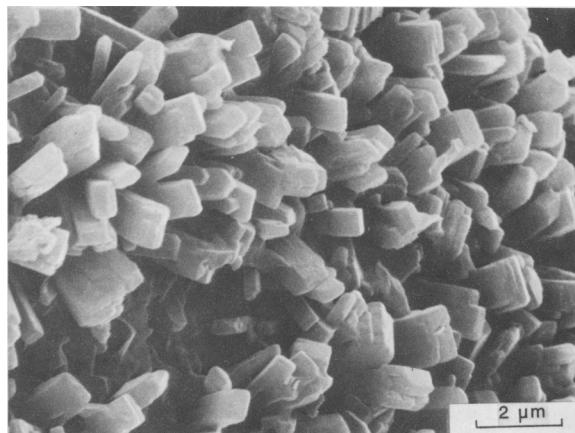


Figure 2. Scanning electron micrograph of tuff from the Big Sandy Formation near Wikieup, Arizona, showing prismatic phillipsite (Table 1, sample 1).

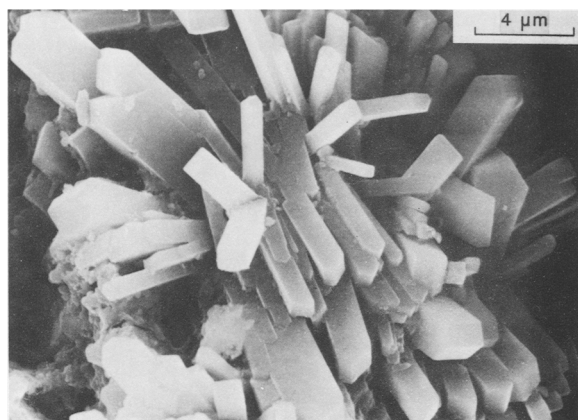


Figure 3. Scanning electron micrograph of tuff from Pleistocene Lake Tecopa near Shoshone, California, showing rosette of prismatic phillipsite (Table 1, sample 2).

not be resolved due to the close overlap and similar intensities of many reflections. All reflections lying within one half the observed full width at half maximum of a peak position and whose calculated intensities were above a minimum threshold set by the weakest observable indexed reflection were considered overlapped.

## RESULTS AND DISCUSSION

### Chemical composition

The chemical composition and unit-cell contents of five phillipsites from saline, alkaline-lake deposits are given in Table 1. As can be seen from Figure 1, these lacustrine phillipsites are siliceous and alkalic, characteristics that set them apart from phillipsites that occur in rocks of other compositions and depositional environments (Sheppard *et al.*, 1970; Passaglia and Vezzalini, 1985). The analyses suggest that  $\text{Fe}^{3+}$  is part of the tetrahedral framework of these phillipsites rather than being present in an impurity. For any phillipsite, the sum of (Si + Al) in the unit cell should equal 16 (half of the oxygens based on a cell of 32 oxygen atoms). For all the phillipsites in Table 1, however, (Si + Al) is less than 16, but is improved to 16 or close to 16 if the  $\text{Fe}^{3+}$  is added. The  $\text{Si}/(\text{Al} + \text{Fe}^{3+})$  ratio is 3.08–3.37, and 75–77% of the framework tetrahedra contain

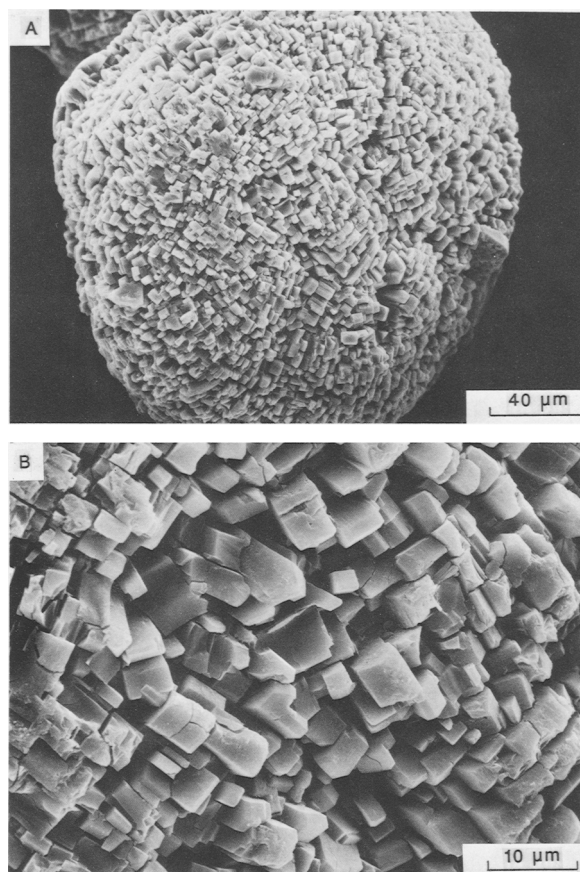


Figure 4. Scanning electron micrograph of spherulitic phillipsite from Pleistocene Lake Tecopa, California (Table 1, sample 3). (A) Spherulite from which adhering clay minerals were removed by ultrasonic treatment prior to being coated with gold; (B) Close-up of exterior of spherulite showing terminations of the phillipsite.

Si. Monovalent exchangeable cations greatly exceed divalent cations, and Na exceeds K, resulting in Na/K ratios of 1.05–3.05.

### Scanning electron microscopy (SEM)

SEM examination of the phillipsite samples from saline, alkaline-lake deposits shows that most are prismatic or spherulitic. Prismatic crystals (Figure 2) are 2–50  $\mu\text{m}$  long and 0.2–5  $\mu\text{m}$  thick and have length-to-

Table 2. Unit-cell parameters and mean indices of refraction for phillipsite<sup>1</sup> from saline, alkaline-lake deposits.

	1	2	3	4	5
$a$ (Å)	9.931 (2)	9.981 (2)	9.985 (5)	9.932 (2)	9.991 (5)
$b$ (Å)	14.151 (3)	14.154 (4)	14.155 (8)	14.150 (3)	14.142 (4)
$c$ (Å)	8.706 (2)	8.690 (3)	8.684 (5)	8.703 (2)	8.650 (6)
$\beta$ (°)	124.57 (2)	124.93 (2)	124.96 (5)	124.57 (2)	125.07 (4)
$V$ (Å <sup>3</sup> )	1007.4 (3)	1000.7 (4)	1006.0 (7)	1007.2 (3)	1000.3 (5)
$n$ (mean)	1.468	1.457	1.461	1.470	1.451

<sup>1</sup> Samples are described in Table 1.

Table 3. X-ray powder diffraction data for phillipsite<sup>1</sup> from Pleistocene Lake Tecopa, California.

h	k	l	Calculated		Observed	
			d <sub>c</sub> (Å)	I <sub>c</sub> (rel)	d <sub>c</sub> (Å)	I <sub>c</sub> (rel)
1	1	0	8.183	8	8.161	14
1	0	$\bar{1}$	8.165	16		
0	0	1	7.118	71	7.104	76
1	1	0	7.085	27		
0	2	0	7.077	59		
1	1	$\bar{1}$	7.073	5		
0	1	1*	6.359	22		
1	2	0	5.353	33	5.347	19
1	2	$\bar{1}$	5.348	20		
0	2	1	5.019	35	5.011	27
2	0	$\bar{1}$	4.992	72	4.974	50
1	0	$\bar{2}$ *	4.282	10	4.285	10
1	1	1	4.105	38	—	—
1	1	$\bar{2}$	4.098	34	—	—
2	2	$\bar{1}$ *	4.080	19	4.084	14
2	1	0*	3.931	5	3.929	8
1	2	1	3.668	4	3.669	6
1	2	$\bar{2}$	3.664	5		
0	1	2	3.451	4	3.453	5
2	3	$\bar{1}$	3.429	5	3.422	3
1	4	0	3.248	30	3.248	41
1	4	$\bar{1}$	3.247	16		
3	0	$\bar{1}$	3.242	16		
3	0	$\bar{2}$	3.239	22		
0	2	2	3.179	59		
1	3	1	3.174	8	3.171	100
1	3	$\bar{2}$	3.171	16		
0	4	1	3.169	100		
3	1	$\bar{1}$	3.161	58		
3	1	$\bar{2}$	3.158	39		
2	3	$\bar{2}$ *	3.087	7	3.096	4
3	2	$\bar{1}$	2.948	20	2.946	19
3	2	$\bar{2}$	2.945	12		
1	4	1	2.730	12	2.734	20
1	4	$\bar{2}$	2.728	15		
3	1	0	2.678	10	2.675	28
2	2	$\bar{3}$	2.679	20		
1	5	0	2.675	22		
2	4	$\bar{2}$	2.674	7		
3	1	$\bar{3}$	2.673	7		
3	2	0*	2.545	5	2.547	4
0	4	$\bar{2}$ *	2.509	12	2.510	4
3	4	$\bar{1}$ *	2.391	14	2.386	5
0	5	$\bar{2}$ *	2.216	6	2.217	2
3	4	$\bar{3}$	2.157	7	2.159	2
3	4	0	2.160	4		
3	2	1	2.071	3	2.067	3
1	6	1	2.067	3		
3	2	$\bar{4}$	2.067	2		
2	2	$\bar{4}$	2.049	5	2.046	3
4	0	0	2.046	5		
1	0	$\bar{4}$ *	1.992	4	1.991	2
5	0	$\bar{3}$	1.977	5	1.975	2
1	1	3	1.974	2		
1	7	0*	1.963	7	1.963	4
3	6	$\bar{1}$	1.908	1	1.904	2
3	6	$\bar{2}$	1.907	4		
5	2	$\bar{2}$	1.905	2		

Table 3. Continued.

h	k	l	Calculated		Observed	
			d <sub>c</sub> (Å)	I <sub>c</sub> (rel)	d <sub>c</sub> (Å)	I <sub>c</sub> (rel)
0	5	3*	1.818	9	1.817	1
4	4	0	1.771	10	1.769	12
0	8	0	1.769	15		
4	6	$\bar{2}$ *	1.715	8	1.716	7
0	6	3*	1.673	7	1.673	2
6	0	$\bar{3}$ *	1.664	14	1.663	5
6	2	$\bar{3}$	1.620	1	1.618	1
6	0	$\bar{4}$	1.620	1		
0	4	4*	1.590	1	1.590	1
6	4	$\bar{2}$	1.474	5	1.473	4
6	2	$\bar{5}$	1.473	3		
6	4	$\bar{5}$ *	1.386	13	1.386	3

\* = Reflection used to constrain first cycle of refinement.

<sup>1</sup> Sample 3 in Tables 1 and 2.

<sup>2</sup> Reflections in brackets are overlapped.

width ratios of about 1.5–10. Some prismatic phillipsite occurs in rosettes (Figure 3) or radial aggregates. Spherulites (Figure 4) are 25–500 μm in diameter and generally show well-developed crystal terminations around their peripheries. Clusters of several mutually interfering spherulites are common, and some of the spherulites are malformed.

#### Optics

These siliceous lacustrine phillipsite samples have parallel or nearly parallel extinction and are length slow. The mean index of refraction is 1.451–1.470 (Table 2), and the birefringence is very low, about 0.002–0.004. Hay (1964) reported a range of 1.438–1.482 for the mean index of refraction of phillipsites from diagenetically altered rhyolitic tuffs of other lacustrine deposits. These values for the mean index are much lower than those generally reported for the phillipsite group (Galli and Loschi Ghittoni, 1972; Gottardi and Galli, 1985, p. 144). The relatively low mean indices of refraction for these phillipsite samples from rhyolitic vitric tuffs of saline, alkaline-lake deposits are undoubtedly due to their relatively high Si and alkali contents, a relationship shown by Galli and Loschi Ghittoni (1972) for the entire phillipsite group.

#### X-ray powder diffraction data

An indexed powder pattern for phillipsite from Pleistocene Lake Tecopa, California, is given in Table 3. Inasmuch as previously published indexed patterns for phillipsite range in the number, position, and intensity of the diffraction peaks (Galli and Loschi Ghittoni, 1972; Gottardi and Galli, 1985), publication seems justified for this representative pattern of the siliceous, lacustrine phillipsites. The approach to multiple indexing on many of the diffraction peaks was described

above. Peak overlap is also indicated by the large observed half widths of many of the diffraction maxima. Attempts to resolve the heavily overlapped maxima, such as those near 3.17 Å, utilizing high-resolution Guinier diffraction, were not successful.

Unit-cell parameters for the five phillipsite samples in Table 1 show the following ranges (Table 2):  $a = 9.931\text{--}9.991$  Å,  $b = 14.142\text{--}14.155$  Å,  $c = 8.650\text{--}8.706$  Å,  $\beta = 124.57\text{--}125.07^\circ$ , and  $V = 1000.3\text{--}1007.4$  Å<sup>3</sup>. Except for a significantly smaller  $b$ , these parameters are within the ranges reported for phillipsite from a variety of rocks and geologic environments (Galli and Loschi Ghittoni, 1972).

Galli and Loschi Ghittoni (1972) found a strong correlation between  $b$  and the Si/(Si + Al + Fe<sup>3+</sup>) ratio for the entire phillipsite group. A decrease in  $b$  was accompanied by an increase in the Si/(Si + Al + Fe<sup>3+</sup>) ratio, and the relationship was expressed by the following regression equation:  $b = 14.87928 - 0.93798 [\text{Si}/(\text{Si} + \text{Al} + \text{Fe}^{3+})]$ . If this equation is solved for the Si/(Si + Al + Fe<sup>3+</sup>) ratio using the measured  $b$  of Table 2, the calculated Si/(Si + Al + Fe<sup>3+</sup>) ratio is 0.77–0.78 for the five siliceous samples. This range, although slightly higher, compares well with the Si/(Si + Al + Fe<sup>3+</sup>) ratio obtained from the chemical analyses, 0.75–0.77 (Table 1). Thus, these values confirm the applicability of Galli and Loschi Ghittoni's regression equation to the Si-rich members of the phillipsite group.

Although a chemical analysis was not available for a phillipsite from a thin rhyolitic tuff in a saline, alkaline-lake deposit at Pine Valley, Nevada (Mumpton, 1984), the measured  $b$  (14.126 Å) was much smaller than any given in Table 2. Applying the regression equation of Galli and Loschi Ghittoni (1972), a Si/(Si + Al + Fe<sup>3+</sup>) ratio of 0.80 was calculated, suggesting that this lacustrine phillipsite is the most siliceous phillipsite to be reported.

All the phillipsites from rhyolitic vitric tuffs in saline, alkaline-lake deposits are predominantly alkalic. Two of the samples in Table 1, however, contain substantial amounts of divalent cations. These same two phillipsites have a significantly smaller  $a$  (Table 2), thus confirming the positive correlation recognized by Galli and Loschi Ghittoni (1972) between  $a$  and the ratio of monovalent cations to total exchangeable cations.

These lacustrine phillipsites owe their distinctive physical properties to their unique chemical composition, namely high Si/Al, (Na + K)/(Na + K + Ca + Mg), and Na/K ratios. Undoubtedly, the principal factors responsible for these particular compositions are the rhyolitic glass parent and the saline, alkaline-lake depositional environment, as previously discussed by Hay (1964) and Sheppard *et al.* (1970).

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