

Wonesite: a new rock-forming silicate from the Post Pond Volcanics, Vermont

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Abstract

A new, Na-rich, trioctahedral layer silicate with the composition $(\text{Ca}_{0.004}\text{Na}_{0.790}\text{K}_{0.145})_{0.939}(\text{Mg}_{4.390}\text{Fe}_{0.778}\text{Mn}_{0.004}\text{Cr}_{0.008}\text{Ti}_{0.074}\text{Al}_{0.620})_{5.874}(\text{Al}_{1.534}\text{Si}_{6.466})_{8.000}\text{O}_{20}(\text{OH},\text{F})_4$ has been discovered in the Post Pond Volcanics, Vermont. The mineral is similar in optical, chemical, and physical properties to phlogopite, the major differences being the high Na/K and smaller d_{001} ($=9.57\text{\AA}$). Monoclinic unit-cell dimensions are $a = 5.312(3)$, $b = 9.163(5)$, $c = 9.825(6)\text{\AA}$, $\beta = 103.18(6)^\circ$ and $V = 465.6(5)\text{\AA}^3$. The mineral coexists with phlogopite, talc, chlorite, cordierite, gedrite, anthophyllite, quartz and plagioclase (An_{10-30}), and is thought to be a stable equilibrium phase under metamorphic conditions. The mineral has been given the name wonesite in honor of David R. Wones.

Introduction

A sodium-rich, trioctahedral layer silicate intermediate in composition between talc and $\text{Na}_2(\text{Mg},\text{Fe})_6\text{Al}_2\text{Si}_6\text{O}_{20}(\text{OH})_4$ has been discovered in the Post Pond Volcanics, Vermont. The mineral is similar optically and chemically to phlogopite but contains an excess of Na over K as the alkali cation. To the authors' knowledge, there are no published reports of a mineral similar in optical, physical, X-ray, or chemical properties. The mineral has been given the name wonesite in honor of David R. Wones, and the name and mineral description have been approved by the International Mineralogical Association. A brief account of this occurrence was given by Spear *et al.* (1978).

Several samples containing wonesite were collected from the Ordovician Post Pond Volcanics in the southwest corner of the Mt. Cube Quadrangle, New Hampshire and Vermont. The samples come from the staurolite-kyanite zone approximately 20 m below the Siluro-Devonian unconformity. Temperature estimates for the metamorphism in this area range from 485° to 535°C (Spear, 1977). The rocks are typically medium-grained (average grain size is approximately 0.5 mm) and include the minerals

quartz, cordierite, gedrite, anthophyllite, chlorite, talc, wonesite, phlogopite, plagioclase (An_{10-30}), rutile, and apatite. The amphiboles (gedrite and anthophyllite) exist as discrete grains and as mutual intergrowths, and some are slightly retrograded to chlorite. Type material has been deposited at the National Museum of Natural History, Washington, D.C., under catalog # NMNH 145724.

Optical, physical, and chemical properties

Wonesite was discovered during routine electron microprobe analysis of biotites from the Post Pond Volcanics, and is closely associated or intergrown with phlogopite, talc, or both. Except where the two minerals are in contact, the mineral is extremely difficult to distinguish from ordinary phlogopite under the petrographic microscope, owing to the similarity in optical properties. Pleochroism in wonesite is strong and is similar to that of coexisting phlogopite: α is pale brown and β and γ are dark brown. In grains that are optically continuous intergrowths of wonesite and phlogopite, the wonesite is slightly paler than the phlogopite (Fig. 1); intergrown talc is much paler than either wonesite or phlogopite.

Textural relations between wonesite and phlogopite (Fig. 1) are very similar to those observed for muscovite-paragonite intergrowths (Eugster *et al.*, 1972, Fig. 7). Typical "bird's-eye maple" extinction, characteristic of nonbrittle micas, is observed in wonesite, and indices of refraction are similar to

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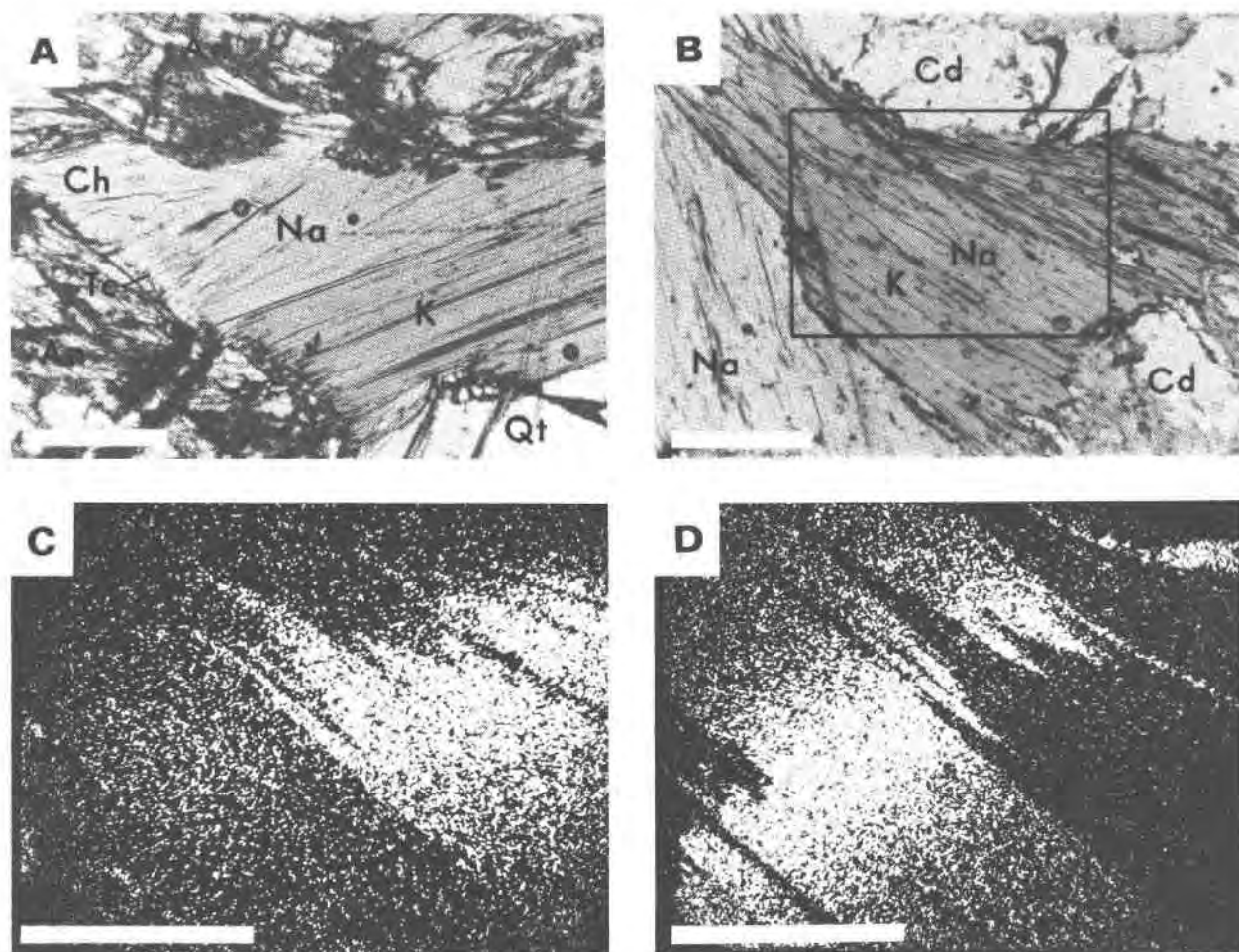


Fig. 1. (A) Photomicrograph of wonesite (Na), phlogopite (K), talc (Tc), chlorite (Ch), anthophyllite (An), and quartz (Qt) taken in plane-polarized light. The intergrown micas in the center of the photograph are optically continuous. Wonesite is slightly paler than phlogopite and darker than talc. (B) Photomicrograph of wonesite (Na), phlogopite (K), and cordierite (Cd) taken in plane-polarized light. The intergrown micas in the center of the photograph are optically continuous; wonesite is slightly paler than phlogopite. The box shows the area of the X-ray photographs C and D. (C) X-ray photograph, Na radiation. Intensity drops off in NW and SE corners of photograph as a result of the spectrometer geometry. (D) X-ray photograph, K radiation. Scale bar is 200 μm .

those of phlogopite: $\beta = \gamma = 1.608 \pm 0.002$ and $\alpha = 1.544 \pm 0.004$, $\delta \approx 0.06$ and $0^\circ < 2V < 5^\circ$.

Single crystals of wonesite are irregular basal-cleavage plates of typical mica habit. Twinning is observed in single-crystal photographs with composition plane (001) and twin axis [310], as is typical with other biotites. Wonesite, phlogopite, and talc form macroscopic epitaxial intergrowths of the (001) plane.

Table 1 gives the chemical compositions of coexisting wonesite, phlogopite and talc, as determined by electron microprobe analysis. All three phases are

relatively homogeneous in composition, both within a single grain and from grain to grain in a single thin section. Traverses across grain boundaries demonstrate that the compositional break between coexisting wonesite and phlogopite is sharp. The Si/Al and Mg/Fe ratios of wonesite are intermediate to those of coexisting talc, which is higher in these ratios, and phlogopite, which is lower. Wonesite is also alkali-deficient, with approximately 1.0 alkali cation per formula unit (out of a possible 2.0 based on 22 oxygens) compared with coexisting phlogopite with 1.4–1.5 alkali cations. The talc also has unusually high

Table 1. Representative electron microprobe analyses of coexisting wonesite, phlogopite, and talc

	Weight % Oxides		
	Wonesite	Phlogopite	Talc
SiO ₂	48.53	40.28	60.76
Al ₂ O ₃	13.73	17.70	2.87
TiO ₂	0.77	0.77	0.06
Cr ₂ O ₃	0.08	0.07	0.0
MgO	22.11	19.61	27.34
FeO*	7.02	8.87	4.25
MnO	0.04	0.04	0.03
CaO	0.04	0.0	0.01
Na ₂ O	3.07	0.50	0.32
K ₂ O	0.85	7.84	0.17**
F†	0.15	0.26	0.11
Total††	96.39	95.94	95.92
Cations based on 22 oxygens			
Si	6.466	5.704	7.795
Al ^{IV}	1.534	2.296	0.205
Al ^{VI}	0.620	0.660	0.228
Ti	0.074	0.079	0.003
Cr	0.008	0.004	0.0
Mg	4.390	4.140	5.227
Fe	0.778	1.047	0.456
Mn	0.004	0.004	0.0
Ca	0.004	0.0	0.0
Na	0.790	0.137	0.075
K	0.145	1.413	0.026
Fe/(Fe + Mg)	0.151	0.202	0.080
	8.000	8.000	8.000
	5.874	5.934	5.914
	0.939	1.550	0.101

*All Fe computed as FeO.

**K₂O content of this talc is anomalously high and may represent contamination with phlogopite. Most talc analyses show 0.02–0.05 wt % K₂O.

†Fluorine analysis by R. Jones.

††Ideal H₂O content for phlogopite is 4.31 wt %, which would bring totals up to approximately 100 wt %.

Al₂O₃ (2.5–3.7 wt. percent) and Na₂O (0.3–0.6 wt. percent) contents.² Wonesite, phlogopite, and talc from these rocks also contain 0.1–0.2 wt. percent fluorine, as determined by electron microprobe analysis (R. Jones, analyst, University of California, Los Angeles).

Infrared spectra were obtained with a Perkin Elmer model 180 spectrophotometer (G. Rossman, analyst, California Institute of Technology). A 21- μ m-thick cleavage fragment was positioned over a 200- μ m-diameter aperture in a metal disk and mounted at the focus of a 6 \times reflecting beam condenser. The convergent nature of the infrared beam at the sample ensured that components of the sample beam were traveling in such directions that vibrational modes of OH⁻ polarized normal to the cleavage fragment could be excited. Absorption in the region of 3593

cm⁻¹ and 3662 cm⁻¹ is interpreted as indicative of the presence of OH⁻ in wonesite. A broad absorption band in the region around 3400 cm⁻¹ may indicate minor H₂O in the interlayer position (George Rossman, personal communication).

The composition of wonesite can be described for the most part in terms of the hypothetical end-member Na₂Mg₆Al₂Si₆O₂₀(OH)₄ and the substitutions K \rightleftharpoons Na, Fe \rightleftharpoons Mg, Al^{VI} + Al^{IV} \rightleftharpoons Mg + Si, \square + Si \rightleftharpoons Na + Al^{IV} and F \rightleftharpoons OH (Figs. 2 and 3). There is a small amount of K \rightleftharpoons Na substitution in wonesite, Na/(Na + K) typically being 0.85–0.95. There is more than 30 percent substitution of the type Al^{VI} + Al^{IV} \rightleftharpoons Mg + Si, so that wonesite has a substantial component of the Na-eastonite end member (see Fig. 3). Another substitution, which results in octahedral vacancies, is 2 \square + Ti⁴⁺ \rightleftharpoons 3R²⁺.

Considerable substitution of the type \square (Interlayer) + Si^{IV} \rightleftharpoons Na + Al^{IV} also occurs in wonesite. This type of substitution is common in the alkali-deficient and silica-rich dioctahedral micas, illite and brammallite. In fact, wonesite might best be described as "trioctahedral brammallite" and the corresponding structural formula is (Na,K)_{1-1.5}(Mg,Fe,Al)₆(Si_{7-6.5},Al_{1-1.5})O₂₀(OH,F)₄ with Na > K and Mg > (Fe + Al). Wonesite, like illite, contains little or no interlayer water, is not penetrated by organic liquids, does not expand irreversibly when heated, and does not swell when treated with glycol. Wonesite, therefore, is both chemically and physically related to the sodic illite brammallite.

The reason for the alkali deficiency in wonesite is not known, but we believe that reported values represent equilibrium alkali contents under metamorphic conditions. If the alkali deficiency were due to leaching from a more Na-rich phase, a complete range of alkali contents would be expected; however, we found that individual grains are relatively homogeneous in composition even where in contact with talc. Moreover, the systematic partitioning of Na, K, and vacancies between wonesite, phlogopite, and talc (Fig. 2) are evidence for the attainment of equilibrium among the alkali cations in these phases.

Crystallography

Unit-cell dimensions determined on single crystals with an automated four-circle diffractometer (MoK α ₁ radiation, λ = 0.70926 Å) are a = 5.312(3), b = 9.163(5), c = 9.825(6) Å, β = 103.18(6)°, and V = 465.6(5) Å³. The crystals are monoclinic, diffraction aspect $C2/m$, Z = 2. The mica is a 1M d polymorph

² Talc analyses with Al₂O₃ and Na₂O values of 2.36 to 3.42 and 0.33 to 0.88 wt. percent, respectively, have been reported by Shreyer and Abraham (1976), who speculated that the alkalis might be contained within the tetrahedral layers in the centers of the (pseudo-) hexagonal rings. Possibly Na may be widely dispersed in the alkali layer of talc. The Al₂O₃ may be accommodated in the talc structure primarily by the substitution Al^{IV}Al^{VI} \rightleftharpoons SiMg.

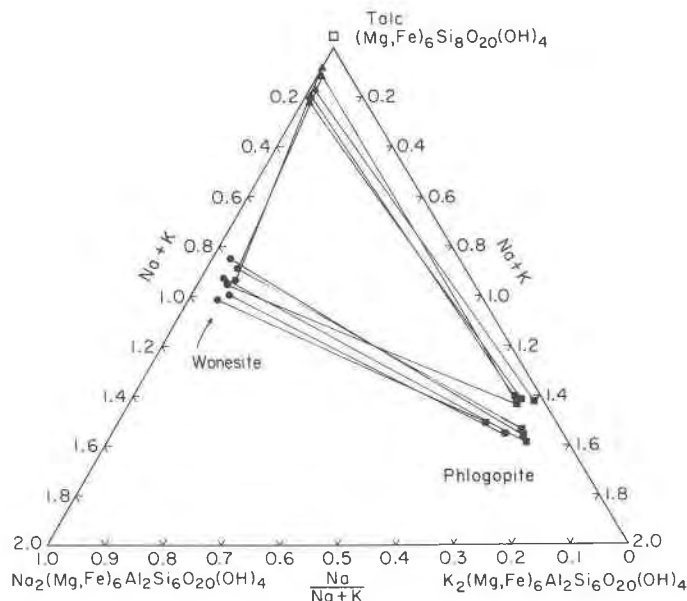


Fig. 2. (A) Compositions of coexisting wonesites (circles), phlogopites (squares), and talcs (triangles) plotted as a function of $\text{Na}/(\text{Na} + \text{K})$ and number of alkali vacancies. The lines connect coexisting phases. Crossing tie lines are believed to reflect analytical uncertainty (error on Na and K = ± 0.05) and the fact that this plot is a condensation of the composition space.

with stacking disorder (typical of illites) present in most specimens. Single-crystal X-ray diffraction photographs have neither streaking nor repeat distances greater than 9.5\AA along c^* . Wonesite, therefore, does not appear to be a mixed-layer or alternating-layer structure with talc and Na-phlogopite layers intermixed on an ordered 1:1 basis.

Powder X-ray diffraction patterns of hand-picked micas from samples containing talc, wonesite, and phlogopite show three distinct peaks in the range $10.4\text{--}9.3\text{\AA}$. Treatment with ethylene glycol had no effect on the positions of these peaks. Similar triplets of peaks appear in the ranges $3.42\text{--}3.07$, $2.56\text{--}2.30$, and $2.05\text{--}1.85\text{\AA}$ and are interpreted as representing the

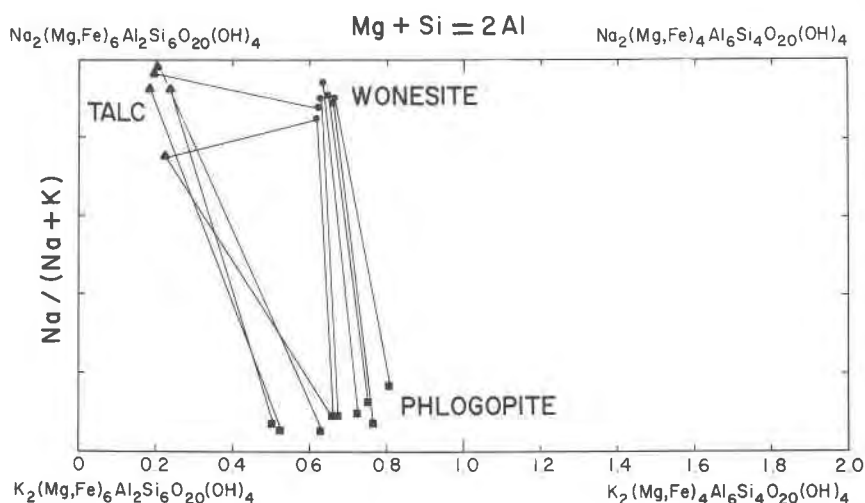


Fig. 3. Compositions of coexisting wonesites (circles), phlogopites (squares), and talcs (triangles), plotted as a function of $\text{Na}/(\text{Na} + \text{K})$ and Tschermak's substitution. The low-Al end-member is phlogopite and the high-Al end-member is aluminous eastonite. Tie lines connect coexisting phases. The one talc analysis that plots at $\text{Na}/(\text{Na} + \text{K}) = 0.75$ is anomalously high in K and may actually plot with the other talc analyses.

001, 003, 004, and 005 interplanar spacings of three distinct layer silicates with d_{001} of 10.08 (phlogopite), 9.62 (wonesite), and 9.35 Å (talc) (± 0.05 Å). The 10.08 Å basal spacing compares well with those reported by Wones (1963) for synthetic biotites, which have a d_{001} ranging from 10.108 to 10.166 Å, and with natural biotites, which have a d_{001} ranging from 9.99 to 10.11 Å (Franzini and Schiaffino, 1963). The 9.62 Å interplanar spacing corresponds to the d_{001} spacing obtained from single crystals of wonesite, 9.57 Å. The smallest d_{001} of 9.35 Å has been assigned to talc (d_{001} of pure talc = 9.31 Å).

Crystal imperfections and intergrowths with phlogopite precluded a complete three-dimensional refinement of the crystal structure of wonesite. It was possible, however, to collect (001) intensity data on one specimen that was epitaxially intergrown with phlogopite. This one-dimensional refinement was analyzed to provide information on the position and electron density of each atom layer parallel to (001). Intensities of all $0 \cdot 0 \cdot l$ reflections, $|l| \leq 14$, were measured on an automated four-circle diffractometer (Finger *et al.*, 1973), using $\text{MoK}\alpha$ radiation. Background counting time and scan rate were adjusted to yield a constant ratio of intensity to its standard deviation ($\sigma_I/I = 0.02$). Intensities were corrected for absorption by the flat plate crystal, $130 \times 130 \times 30 \mu\text{m}$ ($\mu = 15.4 \text{ cm}^{-1}$).

Reflections at $+l$ and $-l$ were averaged; of the 14 reflections, three were unobserved ($l = 12, 13$, and 14) and one (004) had an irregular background, perhaps due to interference with phlogopite diffraction. One-dimensional refinement was completed with the remaining 10 reflections. The first cycles included refinement of a scale factor and the z positional parameters of two oxygen and one tetrahedral cation atomic layers. Interlayer composition was constrained to be 50 percent Na and 50 percent vacancy, and temperature factors were constrained to be equal to those of annite, as observed by Hazen and Burnham (1973). After convergence of positional parameters, all z 's were constrained, and the scale factor, an interlayer occupancy factor, and one temperature factor were refined. The ratios of temperature factors were constrained to be the same as those of annite and phlogopite (Hazen and Burnham, 1973). The resulting model (Table 2) is in agreement with the 10 observed reflections ($wR = 6.3$ percent; $R = 12.5$ percent). One-dimensional refinements of biotites by Franzini and Schiaffino (1963) yielded similar residuals. Calculated and observed structure factors are listed in Table 3.

Table 2. Refined parameters of wonesite; 001 data only*

Atom	x	y	z	B	Occupancy
Na	0	0	0	3(1)	0.55(10)
M1	0	1/2	1/2	0.8(4)	0.86 Mg, 0.14 Fe
M2	0	0.83	1/2	0.8(4)	0.86 Mg, 0.14 Fe
T	0.58	0.17	0.225(1)	0.8(4)	0.80 Si, 0.20 Al
O1	0.82	0.23	0.158(2)	1.7(7)	1.00 oxygen
O2	0.52	0	0.158(2)	1.7(7)	1.00 oxygen
O3	0.63	0.17	0.393(2)	1.2(3)	1.00 oxygen
O4	0.13	0	0.393(2)	1.2(3)	1.00 oxygen

*All x and y parameters were fixed; z parameters of Na, M1 and M2 are constrained by symmetry; occupancies of all sites except Na were fixed; $Z_{O1} = Z_{O2}$; $Z_{O3} = Z_{O4}$; $B_T = B_{M1} = B_{M2} = 0.3B_{Na} = 0.5B_{O1} = 0.5B_{O2} = 0.7B_{O3} = 0.7B_{O4}$. A total of six parameters were refined, including a scale factor, one B , three Z 's and the interlayer occupancy.

One-dimensional structural data on wonesite are consistent with a 50 percent occupancy of the interlayer by Na, as suggested by the electron microprobe analyses. The interlayer thickness (*i.e.* the spacing between adjacent O1–O2 layers = $2 \cdot z_{O1} \cdot c \cdot \sin \beta$) is 3.03 Å, compared with 3.37 Å for biotites. This difference in interlayer thickness is comparable to the difference in interlayer Na and K distances in paragonite and muscovite (Burnham and Radoslovich, 1964). The tetrahedral–octahedral–tetrahedral “sandwich” thickness of wonesite (6.56 Å) is similar to that of many biotites (6.60 Å, Franzini and Schiaffino, 1963). Furthermore, as noted above, the a and b axes of coexisting wonesite and phlogopite are identical. It appears, therefore, that the only major structural and compositional differences between wonesite and phlogopite are associated with the interlayer cation. To a first approximation, the octahedral–tetrahedral–octahedral “sandwich” layers are modules (Thompson, 1978) common to both micas.

Discussion

Two synthetic Na-rich trioctahedral micas are reported in the literature. Carman (1974) synthesized

Table 3. Calculated and observed structure factors for wonesite; 001 data only

	F_{obs}	F_{calc}
001	74.8	-74.6
002	19.4	-21.5
003	132.7	-133.1
005	59.9	61.9
006	47.5	29.4
007	60.0	-59.3
008	35.6	73.6
009	28.2	-29.1
00.10	34.5	31.2
00.11	32.6	-65.8

three different hydrates of Na-phlogopite (10.0, 12.0, and 15.0 Å phases) with the nominal end-member composition of $\text{Na}_2\text{Mg}_6\text{Al}_2\text{Si}_6\text{O}_{20}(\text{OH})_4$. He found that the least-hydrated variety has a maximum thermal stability limit of 990°C at 2.0 kbar $P_{\text{H}_2\text{O}}$, but that this phase was not quenchable. Hewitt and Wones (1975) synthesized a Na analogue of aluminous eastonite, $\text{Na}_2\text{Mg}_4\text{Al}_2\text{Al}_4\text{Si}_4\text{O}_{20}(\text{OH})_4$, which was also reversibly hydrated to 12 and 15 Å phases. Wonesite is intermediate in octahedral Al between the synthetic micas of Carman and of Hewitt and Wones (see Fig. 2). Natural wonesite also contains Fe, displays considerable alkali deficiency, and does not exhibit multiple hydration states.

There are two reported natural occurrences of minerals resembling wonesite in composition. Kulke (1976) described a mica from a metamorphosed evaporite deposit in northern Africa that is very similar in composition to the Na-phlogopite synthesized by Carman ($\text{Ca}_{0.02}\text{Na}_{1.59}\text{K}_{0.06}$) ($\text{Mg}_{5.90}\text{Al}_{0.14}$) ($\text{Al}_{1.92}\text{Si}_{6.08}$) $\text{O}_{20}(\text{OH})_4$. The other occurrence is from a metamorphosed ultramafic complex in the Alps (Keusen, 1972). The composition of this mica (David Bish, personal communication) is very close to that of the Na-aluminous eastonite synthesized by Hewitt and Wones (1975).

Thompson (1978; personal communication) speculated on the existence of Na-phlogopite in nature and suggested that natural examples are rare because (1) bulk compositions necessary for the formation of this phase are rare and (2) Na-phlogopite should weather quickly to vermiculite. Thompson (1978, p. 245, Fig. 5) speculated that Na-phlogopite should coexist with talc, phlogopite, and edenite. In the occurrence described here, talc and phlogopite coexist with wonesite, but no Ca-amphibole is stable with the Na-mica. The stable amphiboles are the Ca-poor orthoamphiboles anthophyllite and gedrite. This difference is probably due to the presence of additional components in the natural example.

Wonesite in these rocks is believed to be a stable equilibrium phase under metamorphic conditions, based on the textures observed in thin section and the systematic partitioning of alkali elements (Fig. 2A,B) between wonesite, phlogopite, and talc. The presence of wonesite in the Post Pond Volcanics can be attributed to the unusual bulk composition. The bulk compositions of the samples reported here are high in Mg/Fe and contain a moderate amount of Na_2O but very little CaO. Additional CaO would probably stabilize first plagioclase and finally hornblende (these phases are found in metavolcanic rocks

only 100 m to the east) rather than wonesite. Specimens containing phlogopite + talc but no wonesite contain substantially more modal plagioclase (15–30 percent of An_{15-20}) than specimens containing wonesite (0–5 percent).

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