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Crystal structure of lizardite-1*T* from Elba, Italy

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ABSTRACT

Euhedral lizardite-1*T* occurs in the Monte Fico quarries, Elba Island, Italy. The composition is $Mg_{2.74}Fe_{0.16}Al_{0.09}Si_{1.93}Al_{0.07}O_5(OH)_4$. Unit-cell parameters, for two crystals, each with space group $P31m$, are $a = 5.338(4)$ and $c = 7.257(6)$ and $a = 5.330(4)$ and $c = 7.269(6)$ Å. The crystal structures were refined to $R_{tot} = 0.034$ and 0.046, using 251 and 849 independent reflections, respectively. The topology determined from previous refinements is confirmed; in particular, the tetrahedral rotation angle is negative ($\alpha = -2.8$ and $-2.7°$), as expected for the 1*T* polytype.

INTRODUCTION

Lizardite, antigorite, and chrysotile are the Mg-rich serpentines (Whittaker and Zussman, 1956), with an ideal composition of $Mg_3Si_2O_5(OH)_4$. Although chemically simple, they are structurally complex; lizardite has an ideal layer topology, whereas antigorite is modulated, and chrysotile is coiled (Wicks and Whittaker, 1975; Wicks and O'Hanley, 1988; Veblen and Wylie, 1993). To fully understand the occurrences of these minerals, it is important to relate the structural variations to conditions of formation. However, current models describing the structures are not satisfactory because of the lack of suitable single crystals for study. For chrysotile, difficulties in obtaining material include fiber intergrowths, polygonalization of layers, polytype intergrowths, and crystallite bending. Some of these problems exist for antigorite also, but additional problems include twinning and polysomatic faulting. Furthermore, because chemical and textural disequilibrium are the rule for these low-temperature phases, intermixes of several serpentines and impurities are common.

Lizardite is the only Mg-rich serpentine with three-dimensional structural information available, albeit for samples containing little Al. Lizardite-1*T* from Val Sissone was studied by Mellini (1982) at room conditions; lizardite-1*T* and lizardite-2*H*1 from Coli, with lower Al contents, were studied by Mellini and Zanazzi (1987); and Mellini and Zanazzi (1989) examined lizardite-1*T* at high pressures. These samples are referred to as VS, C1T, C2H1, VSHP, and CHP, respectively. Because of the relative simplicity of the ideal 1:1 layers in lizardite and the more extensive structural data available, the lizardite structure is the archetype model for the Mg-rich serpentines. However, relatively poor crystallinity of the samples used in these earlier studies produced artifacts. For example, the Si-O1 bond length is unrealistically short in C1T. To avoid specimen-dependent interpretations and to obtain data about the variability of the structure with respect to composition and genetic differences, we report the refinement of lizardite-1*T* from Monte Fico (MF), Elba Island, Italy (Viti and Mellini, 1993). To our knowledge, this locality, which was closed to mining in 1993 and is now partially overgrown, is the largest occurrence of highly crystalline lizardite.

OCCURRENCE AND SPECIMEN DESCRIPTION

Lherzolitic to harzburgitic serpentinites occur in eastern Elba Island, Italy, in the upper second complex and in the lower fourth complex (Duranti et al., 1992). The second complex is exposed for 5 km, from the village of Rio Marina to Monte Arco. Whereas the northernmost sector preserves retrograde serpentinization textures such as bastites and mesh textures, the southernmost sector was weakly thermally metamorphosed.

Fresh, unweathered, retrograde serpentinites are exposed in two large quarries located in the northern sector on two opposite slopes of Monte Fico. The serpentinites, tectonically disturbed, are fractured to large, subrounded blocks several meters thick. Polygonal serpentine or microcrystalline lizardite commonly occurs as a light green skin decorating the block joints and surrounding the dark serpentinite matrix.

Euhedral lizardite occurs within deformed veins, several meters long and a few decimeters thick, along the block joints. The deformed veins consist of mostly euhedral lizardite-1*T* within a minor chrysotile matrix; the assemblage was produced by dynamic serpentinite recrystallization (O'Hanley, 1991), assisted by deformation and shearing along the joints (Viti and Mellini, 1993, and in preparation). The veins are most evident in the northernmost Monte Fico quarry; data given below refer to specimens from this occurrence.

Euhedral lizardite-1*T* occurs as trigonal prisms, typically 0.3 × 0.3 × 0.7 mm in size; trigonal plates are produced by (001) cleavage. Lineation is evident both in hand specimens and in thin sections. Optically, lizardite is pleochroic, light green parallel to and light yellow perpendicular to [001]; length-fast, with low birefringence and sharp, parallel extinction.

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TABLE 1. Crystal and refinement data

	Crystal MFN3-6	Crystal MFN3-1
Size (mm)	0.25 × 0.25 × 0.08	0.25 × 0.25 × 0.30
a (Å)	5.338(4)	5.330(4)
c (Å)	7.257(6)	7.269(6)
V (Å³)	179.1	178.8
Space group	*P*31*m*	*P*31*m*
Polytype	1*T*	1*T*
θ range (°)	2–30	2–40
Measured reflections	621	1665
Independent reflections	251	849
R_{sym}	0.016	0.022
R_w	0.033	0.048
R_{tot}	0.034	0.046

Note: R_{sym} = discrepancy factors among symmetry-related reflections; R_w = weighted discrepancy factors for reflections with $F \geq 5\sigma_F$; R_{tot} = discrepancy factor for the whole data sets.

Chemical compositions are homogeneous from crystal to crystal, with no zoning. Several electron microprobe determinations, by wavelength-dispersive spectrometry (WDS) and energy-dispersive spectrometry (EDS), indicated homogeneous compositions, with average values for MgO = 37.97, SiO_2 = 39.65, Al_2O_3 = 2.86, FeO = 3.97 wt%. If one assumes the ideal of four OH groups, the crystal chemical formula is $Mg_{2.74}Fe_{0.16}Al_{0.09}Si_{1.93}Al_{0.07}O_5(OH)_4$. The formula compares closely with C1T, $Mg_{2.82}Fe_{0.07}Al_{0.09}Si_{1.94}Al_{0.06}O_5(OH)_4$, differing mostly by greater substitution of Fe for Mg in the MF specimen and with VS lizardite, $Mg_{2.79}Fe_{0.12}Al_{0.07}Si_{1.83}Al_{0.17}O_5(OH)_4$, differing mostly by lower substitution of Al for Si in the MF specimen.

The observed specific gravity, measured by the heavy liquid method, was 2.611.

X-RAY AND ELECTRON DIFFRACTION

Rotation and Weissenberg photographs show sharp diffraction peaks, with limited streaking evident. The diffraction symmetry is $\bar{3}m$, and no *c* periodicity other than 7 Å was observed. Electron diffraction and lattice imaging in the transmission electron microscope, using ground specimens and ion-thinned sections, confirmed the results of the X-ray analysis. Thus, only the 1*T* polytype was found in the Monte Fico quarries, either as crystals useful for X-ray diffraction or as electron-coherent domains. Once again, the 1*T* polytype is preferred among many theoretically possible sequences (Bailey, 1988).

Two crystals, MFN3-6 and MFN3-1, were selected for structure refinement, using a Siemens P4 single-crystal automated diffractometer, equipped with graphite-monochromatized MoKα radiation. The unit-cell parameters, refined by least squares for at least 25 reflections, are reported in Table 1. No crystal decay or instrumental drift was observed during data collection. The data were corrected for Lorentz and polarization factors and for empirically determined absorption effects. Structure refinements were performed starting from idealized structure models. As H atoms could not be confidently located in the difference-Fourier map, they were placed in the positions indicated by the VS structure, with no further refinement. Anisotropic thermal motion was used for all the atoms except for the H atoms. Atomic positional and displacement parameters are given in Table 2; observed and calculated structure factors are given in Table 3.[1]

RESULTS AND DISCUSSION

Lattice parameters

The refined values (*a* = 5.338 and *c* = 7.257 Å for MFN3-6; *a* = 5.330 and *c* = 7.269 Å for MFN3-1) compare closely with those for C1T (*a* = 5.325 and *c* = 7.259

TABLE 2. Final atomic positional and displacement parameters for lizardite-1*T* from Monte Fico

	x	*y*	*z*	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
				MFN3-6					
Si	⅓	⅔	0.0699(5)	101(3)	101(3)	71(5)	0	0	50(3)
Mg	0.3326(2)	0	0.4513(5)	99(6)	96(7)	110(8)	0	0(3)	48(7)
O1	⅓	⅔	0.2920	116(9)	116(9)	44(14)	0	0	58(9)
O2	0.5071(5)	0	−0.0099(6)	239(12)	167(13)	93(11)	0	9(8)	84(13)
O3	0.6662(4)	0	0.5869(4)	128(9)	120(10)	73(13)	0	2(6)	64(9)
O4	0	0	0.2990(6)	132(11)	132(11)	57(16)	0	0	66(11)
H3	0.660	0	0.710	*U* = 0.05					
H4	0	0	0.200	*U* = 0.05					
				MFN3-1					
Si	⅓	⅔	0.0704(3)	118(2)	118(2)	153(3)	0	0	59(2)
Mg	0.3324(1)	0	0.4516(3)	127(4)	125(4)	201(5)	0	−5(2)	62(4)
O1	⅓	⅔	0.2920	134(5)	134(5)	134(7)	0	0	67(5)
O2	0.5067(4)	0	−0.0111(4)	218(6)	155(7)	175(7)	0	4(5)	77(7)
O3	0.6658(3)	0	0.5862(4)	145(4)	152(6)	162(6)	0	−3(3)	76(6)
O4	0	0	0.2990(4)	146(6)	146(6)	159(9)	0	0	73(6)
H3	0.660	0	0.710	*U* = 0.05					
H4	0	0	0.200	*U* = 0.05					

Note: anisotropic displacement parameters (× 10^4) are of the form $\exp[-2\pi^2(\Sigma_i \Sigma_j h_i h_j^2 a_i^{*2} a_j^{*2})]$.

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[1] A copy of Table 3 may be obtained by ordering Document AM-94-569 from the Business Office, Mineralogical Society of America, 1130 Seventeenth Street NW, Suite 330, Washington, DC 20036, U.S.A. Please remit $5.00 in advance for the microfiche.

TABLE 4. Selected interatomic distances (Å) and ditrigonalization values

	MFN3-6	MFN3-1	VS	C1T	C2H1	CHP	VSHP
Si-O1	1.612(4)	1.611(2)	1.616	1.577	1.602	1.62	1.60
-O2 × 3	1.647(2)	1.649(1)	1.646	1.651	1.648	1.65	1.65
Av.	1.638	1.639	1.639	1.632	1.636	1.64	1.64
Mg-O3	2.035(3)	2.029(2)	2.021	2.024	2.021	2.05	1.99
-O3 × 2	2.033(3)	2.028(2)	2.026	2.025	2.021	2.04	2.01
-O4	2.091(3)	2.090(2)	2.082	2.069	2.087	2.16	2.24
-O1 × 2	2.124(2)	2.124(1)	2.121	2.139	2.125	2.08	2.12
Av.	2.073	2.071	2.067	2.070	2.067	2.07	2.08
O3-H3	n.d.	n.d.	0.84	1.16	1.07	n.d.	n.d.
O4-H4	n.d.	n.d.	0.79	0.81	1.01	n.d.	n.d.
O2··O3	3.046	3.048	3.03	3.04	3.09	3.01	2.95
α	−2.8°	−2.7	−3.5	−1.7	+6.4	−2.1	−5.1

Note: lizardite-1*T* from Monte Fico (MFN3-6 and MFN3-1), from Val Sissone (VS), and from Coli (C1T) and lizardite-2*H*1 from Coli (C2H1). VSHP and CHP indicate high-pressure refinements for C1T (7 kbar) and VS (12.5 kbar) specimens.

Å: Mellini and Zanazzi, 1987; $a = 5.322$ and $c = 7.273$ Å: Mellini and Zanazzi, 1989) but are significantly shorter for the c parameter in VS lizardite ($a = 5.332$ and $c = 7.233$ Å: Mellini, 1982; $a = 5.335$ and $c = 7.243$ Å: Mellini and Zanazzi, 1989) and for synthetic Al-rich lizardite (Chernosky, 1975).

The compositional difference between VS and C1T ($Si_{1.83}Al_{0.17}$ and $Si_{1.93}Al_{0.07}$, respectively) decreases the c parameter by an amount equal to a 5-kbar increase. Thus, the structural analogy between Al content and increasing pressure holds for the MF specimens ($Si_{1.94}Al_{0.06}$). Conversely, thermal effects in lizardite can be viewed analogously with a structural model approaching the end-member composition, that is, by decreasing the substitution of Al for Si.

Structural topology

MF lizardite-1*T* matches the results obtained for the VS and C1T specimens, with similar or greater precision. In particular, the 1:1 layer shows trigonal symmetry. For a view of the structure, names of atoms, and the definition of positive and negative α values, see the figures in our previously published papers.

The deviation of the tetrahedral sheet from the ideally hexagonal to the ditrigonal arrangement is limited, being $\alpha = -2.8$ and $-2.7°$ in MFN3-6 and MFN3-1, respectively. The distortion was explained previously, assuming that the best H-bonding system might control the movement of the bridging O2 atoms. Thus, lizardite polytypes with no layer-to-layer rotation (e.g., the 1*T* polytype) should be characterized by negative α values. That prediction, already confirmed for the Coli polytypes (where α is negative in the 1*T* but, as expected, positive in the 2*H*1 polytype), is found in the MF specimens.

Buckling of atomic planes is restricted to variations in z height between O1 and O4 by 0.051 Å, both in MFN3-6 and MFN3-1, compared with 0.064 in VS, 0.116 in C1T, and 0.058 Å in C2H1.

Bond geometry

Table 4 reports selected interatomic distances for the seven available refinements. Apart from an unrealistically short Si-O1 value in the C1T structure, the data are internally consistent and reveal common bond patterns.

The $^{[4]}$Si atom always forms one shorter (apical) and three longer (basal) distances (near 1.61 vs. 1.65 Å, respectively, with an average value close to 1.638 Å). This feature contrasts with the data of Jahanbagloo and Zoltai (1968), who reported 1.68 Å for the distance to the apical O atom and 1.65 Å for the distance to the basal O atoms in multilayer Al-rich lizardite. Thus, the basal Si-O2 bonds are nearly invariant, independent from the different tetrahedral compositions. In contrast, the apical Si-O1 bond distance is sensitive to the Al content of the tetrahedron. Furthermore, the tetrahedral sheet increases thickness with increasing Al content, instead of extending laterally. This response differs from conventional wisdom—that misfit is reduced in serpentines by lateral extension of the tetrahedral sheet.

The $^{[6]}$Mg atom is always asymmetrically coordinated,

TABLE 5. Bond strengths for lizardite-1*T*

	Mg			Si			Total *c*		
	MFN3-6	MFN3-1	VS	MFN3-6	MFN3-1	VS	MFN3-6	MFN3-1	VS
O1	0.312	0.312	0.316	1.033	1.036	1.016	1.969	1.972	1.964
O2				0.940	0.935	0.939	1.880	1.870	1.878
O3	0.399	0.403	0.389				1.197	1.207	1.161
	0.397	0.402	0.386						
O4	0.341	0.342	0.343				1.023	1.026	1.029
Total *a*	2.160	2.173	2.136	3.853	3.841	3.833			

Note: values (vu) were calculated according to Brown and Altermatt, 1985.

being systematically shifted toward the outer O3 (OH) plane (2.02–2.03 Å as average bond length) and away from the inner O1 and O4 planes (≥2.10 Å). Minor differences in the octahedral bonding pattern are explained by considering the compositions. Increasing Fe content of the MF specimens determines the magnitude of the asymmetry in distance, with most of the variation affecting the three short Mg-O3 distances but leaving unaffected the long Mg-O4 and Mg-O1 distances.

Bond strengths

Table 5 reports bond-strength calculations (Brown and Altermatt, 1985) performed assuming the ideal serpentine composition. Because the bonding topologies are similar, the bond strengths are also similar between structures. For this reason, only data for MF and VS refinements are given.

Three main anionic levels occur in the structure: the plane of basal O2 atoms, the inner level of O1 and O4 atoms where tetrahedra and octahedra link, and the outer octahedral level containing O3 (OH). Charge balance is fully satisfied for the inner O1 (OH) and O4. The O2 atoms are systematically underbonded by 0.12 vu, and the O3 (OH) atoms are systematically overbonded by 0.20 vu. Thus, the tetrahedral basal O plane is negatively charged, but the adjacent plane across the interlayer is positively charged; a neutral surface (O1 and O4 atoms) occurs inside. The polarization of the 1:1 layer introduces an efficient mechanism of electrostatic layer-to-layer interactions that link together adjacent layers that are otherwise not chemically bonded. Given O2-O3 contact distances of about 3.0 Å, the imbalance is reduced by approximately 0.10 vu (e.g., Ferraris and Ivaldi, 1988). Thus, when O2-H3-O3 H bonding is considered, the result is complete charge balance.

As regards cations, the M-O bond strength systematically sums to approximately 2.16 vu, which is >2, as expected for pure Mg; the T-O bond strength sums to approximately 3.84 vu, which is <4, as expected for pure Si. These values suggest the occurrence of trivalent cations in both tetrahedral and octahedral sites, as recently confirmed by Mössbauer spectroscopy of Fe-bearing lizardite by O'Hanley and Dyar (1993). However, the values do not balance completely. Note that the excess positive charge in three octahedra should be balanced by the deficient positive charge in two tetrahedra. Thus, if the value of 3.84 vu as the charge for Si is accepted, then 2.11 vu, rather than 2.16, should be expected for the Mg site. Note that the tetrahedral imbalance is constant from refinement to refinement, regardless of the different tetrahedral compositions. Values of 3.915 for VS and 3.965 vu for MF would be expected for charge balance after adjusting for the known compositions.

Thermal motion

Atomic displacement parameters often are unreliable because the values are determined by many variables, such as actual thermal movement around the equilibrium position, static or dynamic resonance among crystallochemically equivalent positions, isomorphic replacements and slightly misplaced positions, stacking faults, absorption effects, and refinement procedures.

Table 6. Diagonal components of the atomic displacement tensors for lizardite, normalized to U_{11} = 100 for Si

	U_{11}	U_{22}	U_{33}
		MFN3-6	
Si	100	100	70
Mg	98	95	109
O1	115	115	44
O2	237	127	71
O3	127	119	72
O4	131	131	56
		MFN3-1	
Si	100	100	130
Mg	107	106	170
O1	113	113	113
O2	185	131	148
O3	123	129	137
O4	124	124	135

However, several possibly meaningful trends are noted. Table 6 reports values of the diagonal terms of the displacement tensors, normalized to U_{11} = 100 for the Si atom. Whereas the U_{11} and U_{22} parameters (apart from O2, see below) cluster close to 100, U_{33} varies from refinement to refinement, systematically smaller than 100 in MFN3-6 but close to 140 for MFN3-1.

That variation may be tentatively explained as due to variable lattice imperfections along [001]: the higher the normalized U_{33} value, the lower the coherency of scattered waves along [001] and the greater the lattice discontinuity in that direction. In contrast, constant U_{11} and U_{22} values indicate similar coherency and comparable structural continuity along [100] and [010]. Thus, the lizardite crystals are "infinite" along [100] or [010] but have considerable mosaic texture along [001].

As regards O2, the anomalously large values occurring for U_{11} may be related to the resonance (static or dynamic) from negatively to positively distorted tetrahedral sheets. Such a resonance requires [100] movement, with a shift from $x \approx 0.505$–0.508 to $x \approx 0.495$–0.492, whereas the y and z shift components are zero.

Unfortunately, comparison with previous refinements was fruitless, in that contradictory trends were observed. The disagreement may be due to different means of data collection and processing procedures, and new data, collected in a common and consistent way, are required to settle this point definitely.

CONCLUSIONS

In conclusion, the two new refinements, when compared with the previous five ones, depict an internally consistent pattern.

Crystals of lizardite are rare but exist in different geological environments. Both thermal stability (Jahanbagloo and Zoltai, 1968; Caruso and Chernosky, 1979) and crystal order are promoted by M^{3+} and T^{3+} cations substituting for M^{2+} octahedral and T^{4+} tetrahedral cations, even in the limited amounts present in the Monte Fico or Coli specimens. The mechanism may be related to as increasing linkage by H bonding from layer to layer. Structural parameters that produce poor crystallinity may be related to decreasing three-dimensional connections; indeed, most chemical bonding develops within the 1:1 layer, with almost no layer-to-layer connection.

The overall structure of lizardite may be described as consisting by two submodules: the 1:1 layer itself and the empty space where two adjacent 1:1 layers meet together through interfacing, hexagonally close-packed O atoms, with no cation inside. The internal dimensions of the 1:1 layer show neither major changes after chemical substitution nor increasing pressure, as indicated by the structure determinations from seven different refinements. Instead, the interlayer thickness varies significantly when compositions are changed (Chernosky, 1975) or when the pressure is increased (Mellini and Zanazzi, 1989). As the interlayer thickness decreases, the ditrigonalization of the tetrahedral sheet increases, either in a positive or in a negative way, as determined by the actual stacking sequence.

A similar trend is observed in amesite samples of different compositions (ideal composition, $Mg_2AlSiAlO_5(OH)_4$: Hall and Bailey, 1979; Anderson and Bailey, 1981; Wiewiora et al., 1991). As the interlayer separation decreases from 3.05 Å of lizardite to 2.74 Å of amesite, α increases from 2–3 to 15°.

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