ANTIGORITE WITH A LARGE SUPERCELL FROM SAGANOSEKI, OITA PREFECTURE, JAPAN

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ABSTRACT

Variations in supercell parameters and chemical compositions for nine samples of antigorite from the Saganoseki area, eastern Kyushu, Japan, were studied by electron diffraction (SAED), X-ray powder diffraction (XRD), electron microprobe (EPMA) and analytical electron microscopy (AEM). The XRD patterns give various A parameters of the supercell (A in the range 42.4–53.6 Å) as well as varying subcell dimensions ($5.41 \leq a \leq 5.44$, $9.24 \leq b \leq 9.26$, $7.26 \leq c \leq 7.28$ Å, $91.0 \leq \beta \leq 91.3°$). Furthermore, SAED patterns show a wider range of A parameters, from 32 to 60 Å ($5.9 \leq M \leq 11.1$, $M = A/a$). The antigorite from Saganoseki has a high Al/Fe value. The large parameters of the supercell in antigorite (M between 8.5 and 10.0) are characteristic of serpentinites formed under low-temperature and high-pressure (pumpellyite–actinolite facies) conditions. The AEM results suggest that the correlations of the Fe contents or the Al contents with M parameter are not always distinct within a specimen, and may depend on other structural and chemical parameters.

Keywords: antigorite, superstructure, supercell, serpentine, crystal chemistry, X-ray diffraction, electron diffraction, analytical electron microscopy.

SOMMAIRE

Nous avons étudié les variations dans la surstructure et la composition chimique de neuf échantillons d'antigorite provenant de la région de Saganoseki, dans la partie orientale de Kyushu, au Japon, par diffraction d'électrons, diffraction X, microsonde électronique et microscopie électronique analytique. Les clichés de diffraction indiquent une variation du paramètre A de la surstructure, entre 42.4 et 53.6 Å, ainsi que dans les paramètres de la sous-maille (a entre 5.41 et 5.44 Å, b entre 9.24 et 9.26 Å, c entre 7.26 et 7.28 Å, β entre 91.0 et 91.3°). De plus, la diffraction d'électrons dans une aire restreinte montre une grande variabilité dans le paramètre A, entre 32 et 60 Å ($5.9 \leq M \leq 11.1$, $M = A/a$). Les échantillons d'antigorite de Saganoseki possèdent un rapport Al/Fe élevé. Ceux dont la surstructure est plus grande ($8.5 \leq M \leq 10.0$) seraient typiques d'un milieu de croissance à faible température et à pression élevée (facies pumpellyite–actinote). D'après les résultats d'analyses en microscopie électronique à transmission, les corrélations possibles entre M et la teneur en Fe ou en Al dans un échantillon ne sont pas évidentes dans tous les cas, et pourraient résulter d'autres facteurs structuraux et chimiques.

(Traduit par la Rédaction)

Mots-clés: antigorite, surstructuré, surmaille, serpentine, chimie cristalline, diffraction X, diffraction d'électrons, microscopie électronique analytique.

INTRODUCTION

Antigorite is characterized by a superstructure with large periodicity along the X direction based on an alternating-wave structure (Kunze 1956, 1958). The large A parameter was first shown to be 43.5 Å (Aruja 1945). Further studies have shown that antigorite does not have a fixed composition, but forms a series of variable compositions and structures (Kunze 1961, Uehara & Shirozu 1985, Spinnler 1985). Kunze (1961) first proposed a series of discrete compositions

that are chemographically colinear with chrysotile and lizardite, $Mg_3Si_2O_5(OH)_4$, and talc, $Mg_3Si_4O_{10}(OH)_2$. The compositional complexity is mostly due to the occurrence of different periodicities of the superstructure in antigorite, but the occurrence of such superstructures in nature has remained poorly documented.

Mellini *et al.* (1987) found that increasing temperature of formation can be correlated with a shorter periodicity of the supercell in the antigorite serpentines of the Swiss and Italian Alps. Uehara & Shirozu (1985) investigated the variation of supercell in antigorite from Nishisonogi and Sasaguri; the Nishisonogi antigorite shows a larger supercell than the Sasaguri antigorite, with identical Fe^{2+} contents. Also, cell dimensions of antigorite from each locality show a different relationship to chemical composition.

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The reported values of A obtained by electron diffraction are 33.7–43.1 Å (Zussman *et al.* 1957), 25.7–51.5 Å (Kunze 1961), and 32.7–51.4 Å (Uehara & Shirozu 1985). A very long superstructure group with A between 80 and 110 Å (Brindley *et al.* 1958), and a short superstructure with A in the range 16–19 Å (Chapman & Zussman 1959) also were recorded.

Mellini & Zussman (1986) showed the 19 Å and 16 Å periodicities given by Chapman & Zussman (1959) for "picrolite" from Taberg do not pertain to antigorite, but rather to the 18 Å $d(200)$ interplanar spacing of carlosturanite S_5X and the 15.5 Å $d(100)$ of the S_4X polysome, respectively.

In this paper, A is used for the periodicity of the supercell along the X axis of antigorite, and a is used for the subcell X axis periodicity, about 5.4 Å. Uehara & Shirozu (1985) have shown that 1) powder patterns of antigorite with various supercell parameters can be indexed by using the M parameter ($M = A/a$), and 2) the supercell parameters are closely related to chemical composition.

Ultramafic rocks occurring in Saganoseki, eastern Kyushu, Japan, generally consist of serpentinized dunite and lherzolite, that have been affected by low-temperature regional metamorphism. Antigorite with a supercell parameter of about 53 Å were found in the Saganoseki serpentinites. In this paper, the mineralogical properties of the Saganoseki antigorite are discussed and compared with those of antigorite from the Sasaguri and Nishisonogi serpentinites (Fig. 1; Uehara & Shirozu 1985, Uehara 1987) and those of other examples of antigorite in the literature.

SERPENTINE MINERALS
IN THE SAGANOSEKI DISTRICT

The Sanbagawa metamorphic rocks of the Saganoseki Peninsula, eastern Kyushu, mainly consist of low-grade pelitic, psammitic and siliceous schists and serpentinites. The area is the western end of the Sanbagawa belt, which is a typical high-pressure metamorphic terrane in the Japanese Islands (*e.g.*,
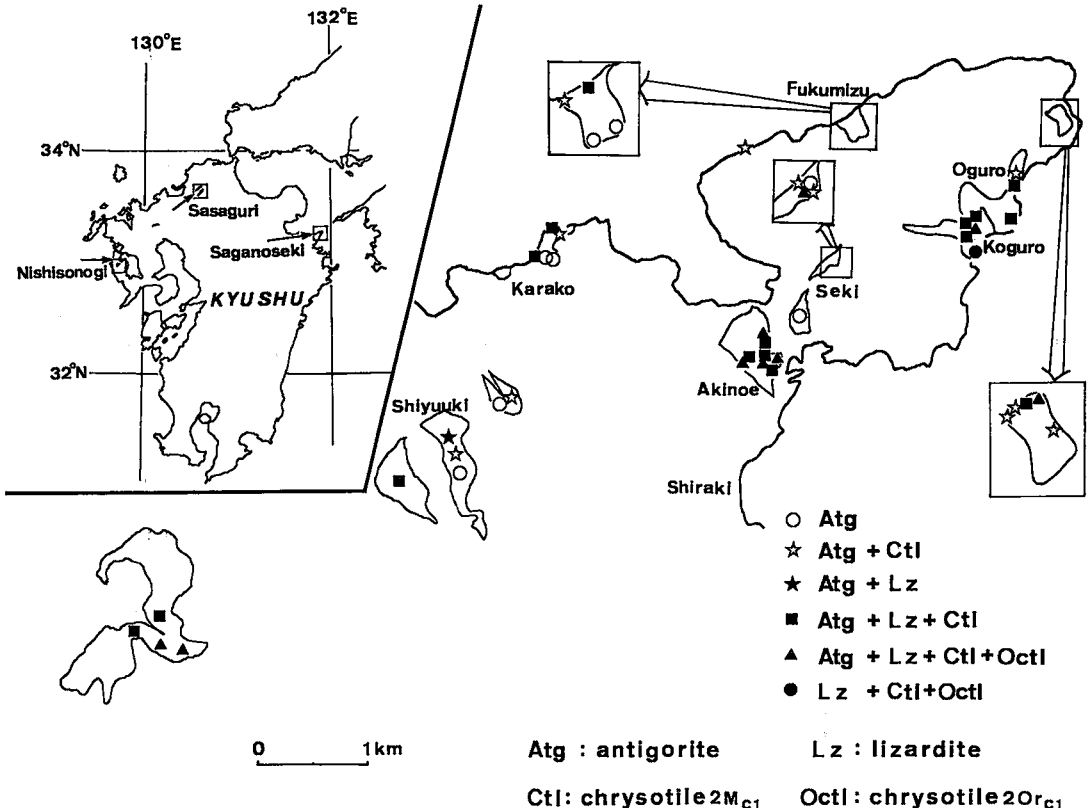

FIG. 1. Distribution of serpentine minerals in the Saganoseki serpentinites.

Banno 1986). The pumpellyite–actinolite-facies metabasites of the Sanbagawa belt are estimated to have recrystallized at 250–330°C and pressures of 3 to 5.2 kbar (Nakajima *et al.* 1977, Nakajima, 1983).

Geological studies of the Saganoseki metamorphic rocks were reported by Yamamoto & Teshima (1971), Yoshikawa *et al.* (1980), Sonoda (1985) and Ikeda (1987). The geological structure of this area is characterized by the Furumiya Anticline (Yoshikawa *et al.* 1980), whose axis trends east–northeast. Serpentinites occur as large massive bodies along the axis of the Furumiya Anticline, and also as small sheared bodies in the northern flank of the Furumiya Anticline. In the eastern part of the Saganoseki Peninsula, serpentinite bodies with metagabbro are distributed along a syncline in the crystalline schist.

In the basic schist and metagabbro, pumpellyite and actinolite are commonly formed, and lawsonite + pumpellyite + quartz + albite + chlorite + calcite and glaucophane are found to coexist (Ikeda 1987), so that the metamorphic facies of this region is pumpellyite–actinolite of relatively high-pressure type.

The serpentinites in this district are generally massive or schistose, and green to dark in color. Under the optical microscope, they are fine-grained, granular or fibrous, and have a mesh texture or bastite texture consistent with pseudomorphism after orthopyroxene or clinopyroxene. A preliminary analysis to investigate the mineral composition of serpentinites was undertaken by X-ray powder diffraction. Identification of the serpentine species, antigorite, lizardite and chrysotile, was mainly done by comparison with the powder data of Whittaker & Zussman (1956). The nomenclature of chrysotile polytypes follows the notation of Wicks & Whittaker (1975).

The constituent minerals of the Saganoseki serpentinites are identified to be antigorite, lizardite, chrysotile, chlorite, tremolite, diopside, calcite, dolomite, magnesite, magnetite and pyrite. Talc and brucite have not been found in the serpentinites. The distribution of rock-forming serpentine minerals is shown in Figure 1. In this figure, the macroscopic vein-forming serpentines were excluded.

Most bodies of serpentinite consist mainly of antigorite with variable amounts of chrysotile and lizardite. Nine samples from Saganoseki that consist of antigorite – chrysotile $2M_{c1}$ – lizardite were selected for this study, mainly on the basis of variation of X-ray powder patterns. Two samples (SG6, SG1) consist of antigorite with a small amount of lizardite or chrysotile, three samples (SG21B, SG26B, SG48A) consist mainly of antigorite, and the other four samples (SG12, SG16, SG49, SG80) consist of relatively smaller amounts of antigorite than the former five samples (Table 1). The mineralogical properties of these samples of antigorite were studied by X-ray powder diffraction (XRD), selected-area electron diffraction (SAED), electron-probe microanalysis (EPMA), and analytical electron microscopy (AEM).

TABLE 1. SAMPLES OF ANTIGORITE ROCKS FROM SAGANOSEKI

Sample	Color and texture (Locality)	Atg	Ctl	Lz	Chl	Di	Mag	Py
SG6	Black brecciated mass with green fibrous parts (Seki)	+++	+				+	
SG1	Grayish green massive with bastite (Seki)	+++		+			+	
SG21B	Dark green massive with fine chrysotile vein (Shiyuuki)	+++					+	
SG26B	Dark green massive (Shiyuuki)	+++				+	+	
SG48A	Dark green massive with bastite & chrysotile vein (Oguro)	+++			+	+	+	
SG12	Black massive (Karakou)	++	+	++			+	
SG16	Black massive (Karakou)	++	+	++			+	
SG49	Black to dark green massive (Oguro)	+++	++	++			+	+
SG80	Black massive (Akinoe)	+++	++	++	+		+	

*:relative amounts were estimated by XRD.

Atg:antigorite. Ctl:chrysotile. Lz:lizardite. Chl:chlorite. Di:diopside. Mag:magnetite: Py:pyrite.

+++:large amount. ++:medium. +:small.

Experimental Method

X-ray powder-diffraction patterns were recorded with a Rigaku RAD–IIA diffractometer using filtered CuKα radiation and a standard aluminum sample holder. Silicon powder was used as an internal standard.

Electron optical observations were made with JEOL JEM–200B and JEM–2000FX electron microscopes in the HVEM laboratory of Kyushu University. The pulverized materials were disaggregated ultrasonically and mounted on copper grids. The JEM–200B microscope, which was mainly used for obtaining selected-area electron-diffraction patterns, was operated at 200 kV; a selected-area aperture of 10 μm diameter was used for electron diffraction, which has a projected diameter on the specimen of about 0.2 μm.

Chemical compositions of antigorite were determined with a JEOL–JXA 733 electron microprobe operated at 15 kV accelerating voltage and a diameter of the electron beam of 1 μm. The following standards were used: synthetic $CaSiO_3$ (Si,Ca), corundum (Al), hematite (Fe), periclase (Mg), NiO (Ni), MnO (Mn), and TiO_2 (Ti) and natural chromite (Cr). Electron-microprobe data using wavelength dispersion were ZAF-corrected, total Fe was considered as FeO, and H_2O was estimated by difference.

Some analyses were performed with a JEOL model JEM–2000FX analytical electron microscope. The AEM was used to analyze the particle area corresponding to that used to obtain selected-area electron-diffraction patterns in order to determine the value of M. The corrections were made by the ratio method of Cliff & Lorimer (1975). Sample A1 (Uehara & Shirozu 1985) was used as the standard material for experimental measurement of the k factor. The JEM–2000FX was operated in the TEM mode with a beam diameter of 0.4 μm at an accelerating voltage of 200 kV. More detailed procedures are given by Uehara et al. (1991).

Results

X-ray powder patterns

An indexing of the powder patterns of antigorite was first performed by Whittaker & Zussman (1956) using a supercell A of eight units of the subcell a. Uehara & Shirozu (1985) have re-examined the powder patterns of various samples of antigorite and re-indexed these using the M parameter, which is the ratio of the supercell A parameter to the subcell a parameter (A/a).

The antigorite samples from Saganoseki generally show broadened reflections attributed to the supercell [(h±u)/M, k, l; u ≠ 0] compared with the Sasaguri and Nishisonogi samples (Uehara & Shirozu 1985). This may be caused by a heterogeneous distribution of dif-

TABLE 2. CELL PARAMETERS OF ANTIGORITE FROM SAGANOSEKI

	SG6	SG1	SG21B	SG26B	SG48A	SG49
$a(\text{Å})$	5.410	5.428	5.407	5.418	5.414	5.437
$b(\text{Å})$	9.244	9.239	9.240	9.251	9.244	9.258
$c(\text{Å})$	7.280	7.277	7.277	7.268	7.267	7.263
$β(°)$	91.07	91.33	91.08	91.03	91.12	91.10
M	9.90	8.80	9.65	9.56	9.12	7.80
$A(\text{Å})$	53.56	48.20	52.18	51.80	49.37	42.41

The symbols for most data-points in Figure 5 exceed a 1σ error bar in size; the error expected in M is about ±0.5.

ferent parameters of the supercell in a sample or small size of crystallites. Table 2 shows the cell parameters of six samples refined by a least-squares method using the reflections with d greater than 1.53 Å. The cell parameters of the other three samples were not refined, because of the breadth of the reflections or the presence of chrysotile and lizardite. The ranges of cell parameters and M values obtained for the Saganoseki materials are: 5.407 ≤ a ≤ 5.437 Å, 9.240 ≤ b ≤ 9.258 Å, 7.263 ≤ c ≤ 7.277 Å, 91.07 ≤ β ≤ 91.33°, 42.4 ≤ A ≤ 53.6 Å, 7.8 ≤ M ≤ 9.9. In general, the a and β parameters are smaller, and the A and M parameters are larger, than those of the Sasaguri and Nishisonogi material (Uehara & Shirozu 1985). The reason for variations in these parameters will be discussed in the last section.

Electron-diffraction patterns

The supercell parameters were measured by selected-area electron-diffraction patterns (Fig. 2). The supercell A parameters corresponding to the intervals of the clustered spots on the electron-diffraction patterns are within a range from about 32 Å to 60 Å (5.9 ≤ M ≤ 11.1). Many nonintegral types of superstructures (M ≠ (2n + 1)/2, where n is an integer) were found in the Saganoseki antigorite. Variations in the values of M in a single specimen were examined in all samples. The results, given in Figure 3, show that the differences in the M value are mostly within ±1.2, but rarely as large as ±1.5. The differences are the same as those of the Sasaguri and Nishisonogi samples (Uehara & Shirozu 1985). Therefore, the broad supercell-related reflections in X-ray powder patterns are probably mainly caused by the small size of the crystallites.

The large M values (between 9.5 and 11.1) obtained from these samples by SAED have not commonly been reported elsewhere. Antigorite with large parameters of the supercell of more than 51 Å (M = 9.5) had only been found in one sample described by
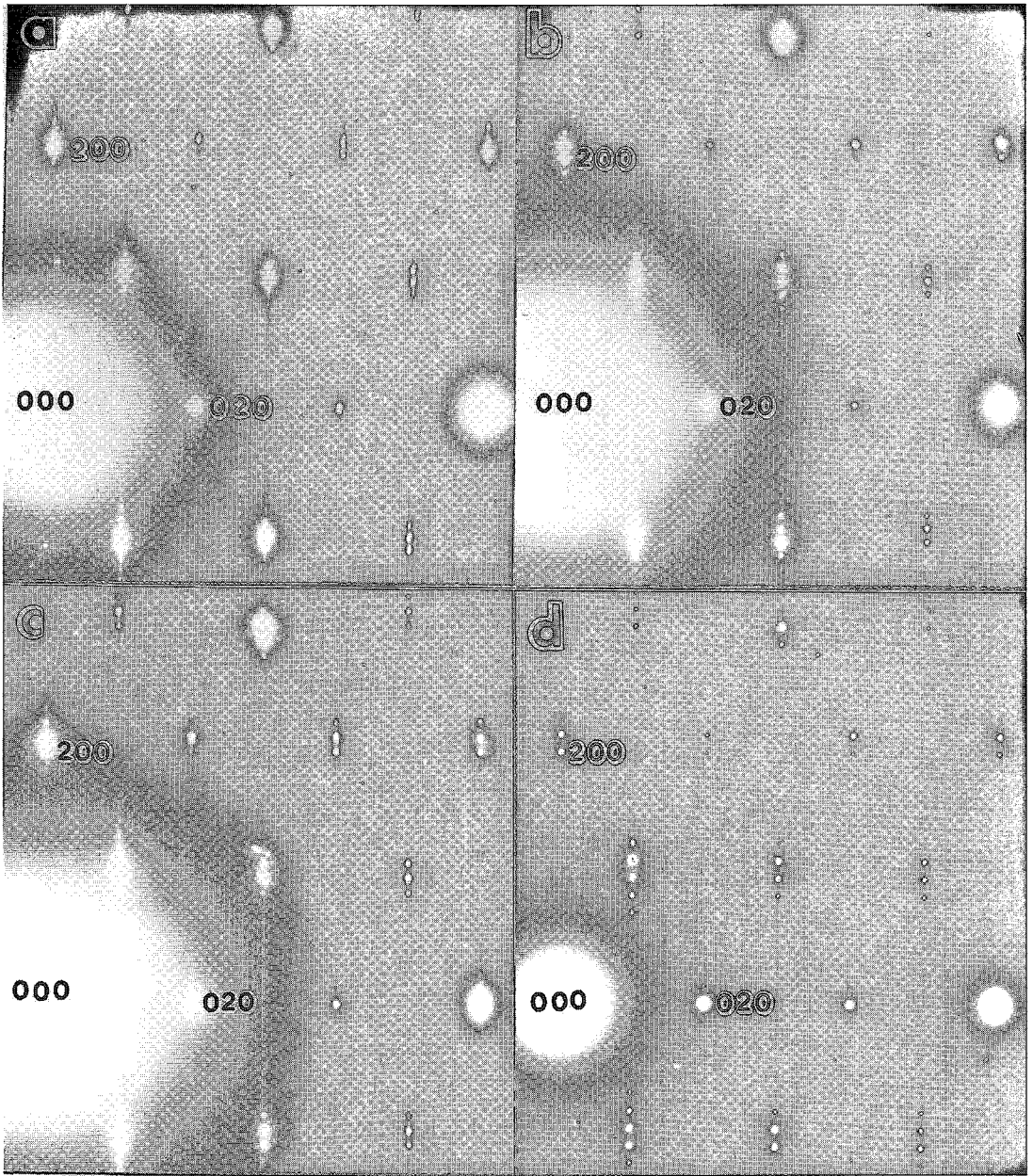
FIG. 2. Electron-diffraction patterns of antigorite from Saganoseki. a) $M = 10.2$ (SG6), b) $M = 9.0$ (SG48A), c) $M = 8.5$ (SG1), d) $M = 7.0$ (SG80).

Kunze (1961), the "Yu Yen Stone" sample (Zussman *et al.* 1957), and a few crystals in some Nishisonogi and Sasaguri samples (Uehara & Shirozu 1985) and D1335 (Mellini *et al.* 1987).

Chemical composition

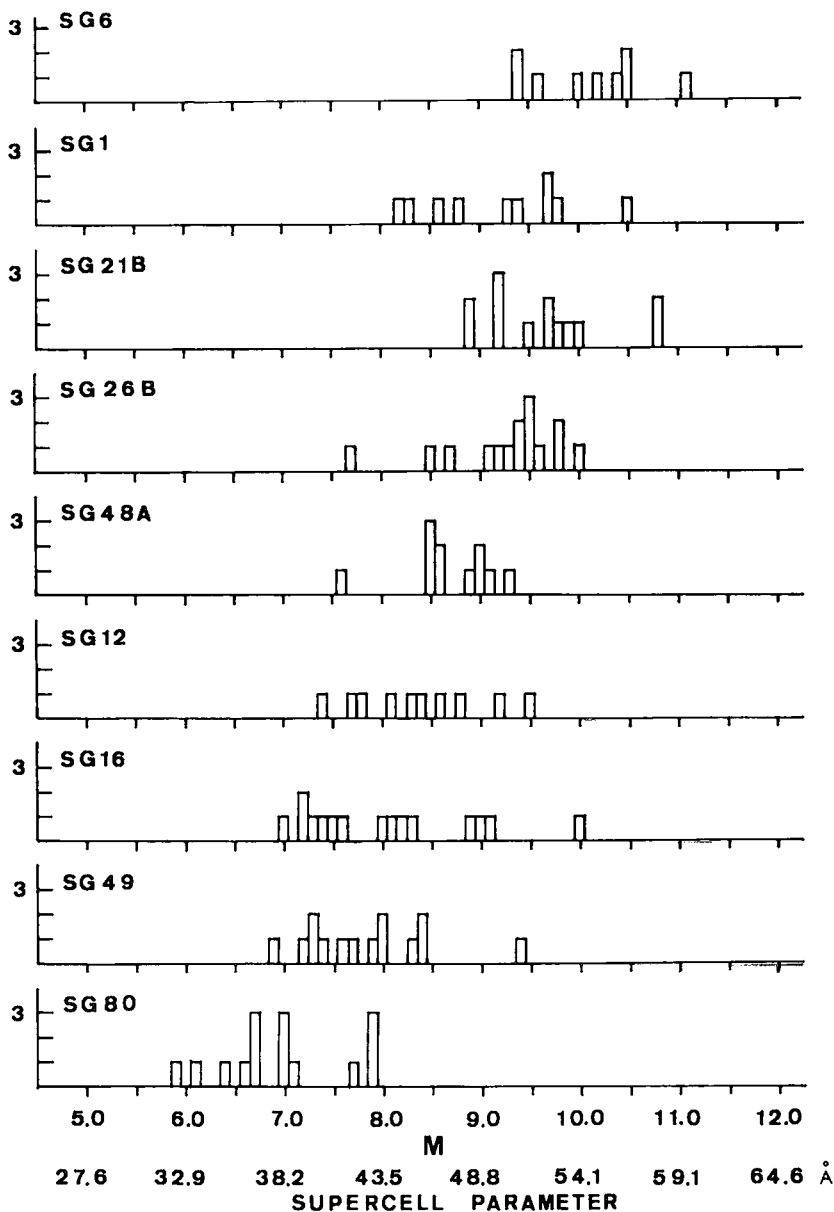Electron-microprobe data were obtained from six samples for which M values could be measured by

FIG. 3. Histogram of M values in the antigorite specimens from Saganoseki, as measured by selected-area electron diffraction.

XRD. The compositions reported in Table 3 are averages of seven or more readings. The samples of Saganoseki antigorite examined are chemically more or less heterogeneous, particularly in Al and Fe contents. The variation is occasionally over 2 wt% in Al_2O_3 in sample SG49, and in ΣFeO (total iron expressed as Fe^{2+}) in sample SG1.

Uehara & Shirozu (1985) showed that the formula for subcell composition having a variable M is $Mg_6Si_{4(1+1/2M)}O_{10(1+1/2M)}(OH)_{8-2/M}$. The cation proportions of the antigorite analyzed, which were calculated on the basis of $(14+4/M)$ atoms of oxygen, also are presented in Table 3. The sum of octahedrally coordinated cations is slightly greater than six. This

TABLE 3. RESULTS OF ELECTRON-MICROPROBE ANALYSES OF ANTIGORITE FROM SAGANOSEKI

	SG6(n=9)	SG1(n=9)	SG21B(n=9)	SG26B(n=7)	SG48A(n=9)	SG49(n=7)
SiO₂	43.25(0.31)	42.16(0.95)	43.73(0.32)	42.36(0.36)	42.98(0.71)	42.07(0.83)
Al₂O₃	1.20(0.26)	2.00(0.78)	0.62(0.11)	2.26(0.31)	1.91(0.47)	3.30(1.16)
Cr₂O₃	0.18(0.08)	0.16(0.08)	0.24(0.11)	0.04(0.04)	0.19(0.24)	0.27(0.06)
FeO*	2.53(0.17)	4.99(1.03)	2.01(0.17)	4.64(0.21)	3.30(0.23)	5.67(0.11)
MgO	41.35(1.20)	39.15(1.11)	41.79(0.32)	38.96(0.49)	40.74(0.59)	37.93(0.40)
NiO	0.10(0.10)	0.17(0.12)	0.06(0.07)	0.19(0.06)	0.05(0.04)	0.05(0.05)
MnO	0.08(0.06)	0.09(0.07)	0.07(0.04)	0.12(0.04)	0.09(0.03)	0.10(0.06)
CaO	0.02(0.02)	0.02(0.02)	0.01(0.01)	0.01(0.01)	0.01(0.02)	0.03(0.02)
TiO	0.01(0.02)	0.04(0.03)	0.03(0.03)	0.04(0.06)	0.02(0.03)	0.04(0.04)
Total	88.70	88.78	88.74	88.62	89.30	89.46
H₂O**	11.30(1.08)	11.22(0.36)	11.26(0.72)	11.38(0.51)	10.70(0.43)	10.54(0.30)

Cation numbers for antigorites on the basis of $(14+4/M)$ oxygens***

Si	4.078	4.039	4.120	4.043	4.050	4.022
Al	0.124	0.188	0.069	0.166	0.169	0.234
sum	4.202	4.227	4.189	4.209	4.219	4.256
$4(1+1/2M)$	4.202	4.227	4.207	4.209	4.219	4.256
Al	0.009	0.038	0.000	0.088	0.043	0.138
Cr	0.013	0.012	0.018	0.003	0.014	0.020
Fe	0.199	0.400	0.159	0.371	0.260	0.453
Mg	5.813	5.592	5.870	5.544	5.723	5.406
Ni	0.007	0.013	0.005	0.015	0.004	0.004
Mn	0.007	0.008	0.005	0.009	0.007	0.008
Ca	0.002	0.002	0.001	0.001	0.001	0.003
Ti	0.000	0.003	0.002	0.004	0.001	0.003
sum	6.131	6.068	6.064	6.034	6.053	6.035
M =	9.90	8.80	9.65	9.56	9.12	7.80

*: total Fe as FeO.   **: estimated by difference.   ***:calculated as anhydrous.
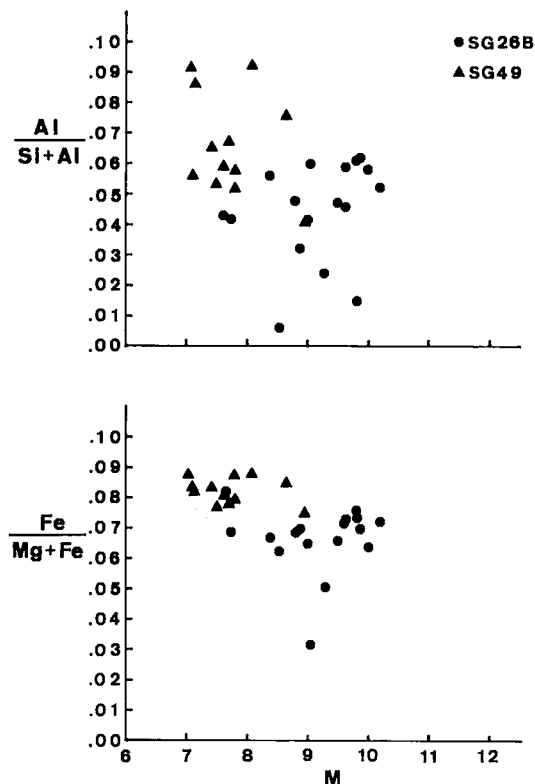n: numbers of analysed points. Values in parentheses are one standard deviation.

FIG. 4. Values of M versus Fe/(Mg + Fe) and Al/(Si + Al) ratios obtained by analytical electron microscopy.

discrepancy may be caused by systematic errors in the electron-microprobe analyses, or chrysotile contamination at the points analyzed.

Analytical electron microscopy

Antigorite specimens commonly are composed of different polysomes with different parameters of the supercell (Fig. 3). There is a possibility that the chemical composition obtained by EPMA represents not a single phase, but a mixture of polysomes.

The AEM was used for two specimens (SG26B and SG49) to analyze the particle areas corresponding to those used to obtain selected-area electron-diffraction patterns for measurement of the M value. The M values are plotted against Al/(Si+Al) and Fe/(Mg+Fe) in Figure 4. The correlations between the Fe contents or the Al contents and the M parameters are not distinct. This suggests that antigorite of different superstructure can have similar Fe and Al contents, and also that antigorite with similar superstructures can have different Fe and Al contents in a specimen.

DISCUSSION AND CONCLUSIONS

Supercell parameter and subcell parameters

The number of subcells contained in a wavelength of alternating-wave structure, which corresponds to the M value, is perhaps the most important parameter in a series of antigorite samples with different super-
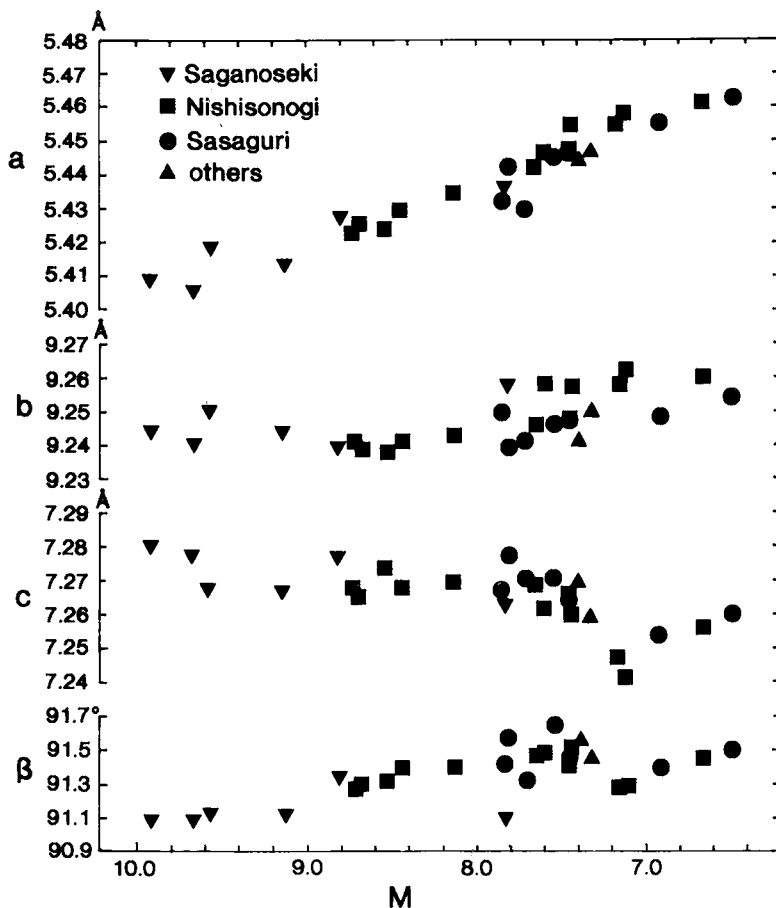
Fig. 5. Cell dimensions *versus* M values for the samples of antigorite.

structures. The antigorite in the Saganoseki metamorphic rocks are characterized by the presence of large parameters of the supercell ($9.6 \leq M \leq 10$).

The cell parameters obtained here by XRD (Table 2) and those previously reported for antigorite (Uehara & Shirozu 1985, Table 4) are plotted against M value in Figure 5. With decreasing M value, the a, b, and β parameters increase, whereas c decreases. The relationships are, however, not well defined, except for the a parameter.

The following linear correlation between M and a was obtained using the above twenty-eight sets of cell parameters:

$$M = 303.84 - 54.41a \qquad (1)$$

Its correlation coefficient is –0.96. Since M is A/a, the A parameter is obtained by $a \times M$, and so the relationship between A and M can also be expressed as follows:

$$A = 1.132 + 5.293M \qquad (2)$$

or

$$M = (A - 1.132)/5.293 \qquad (3)$$

Cation substitution

In antigorite, the incorporation of trivalent cations Al^{3+}, Fe^{3+} and Cr^{3+} for Mg or Si, and of the divalent cations Fe^{2+}, Ni^{2+} for Mg, can occur. Substitutions of Si and Mg by Al^{3+}, Fe^{3+} and Fe^{2+} are most common. Generally, Fe^{2+} tends to dominate over Fe^{3+} in the specimens described in previous papers (Faust & Fahey 1962, Whittaker & Wicks 1970, Uehara & Shirozu 1985). Figure 6 shows the Al_2O_3 and ΣFeO (total iron as Fe^{2+}) contents of the Saganoseki antigorite, together with some data obtained by wet-chemical methods (Hess *et al.* 1952, Zussman 1954,
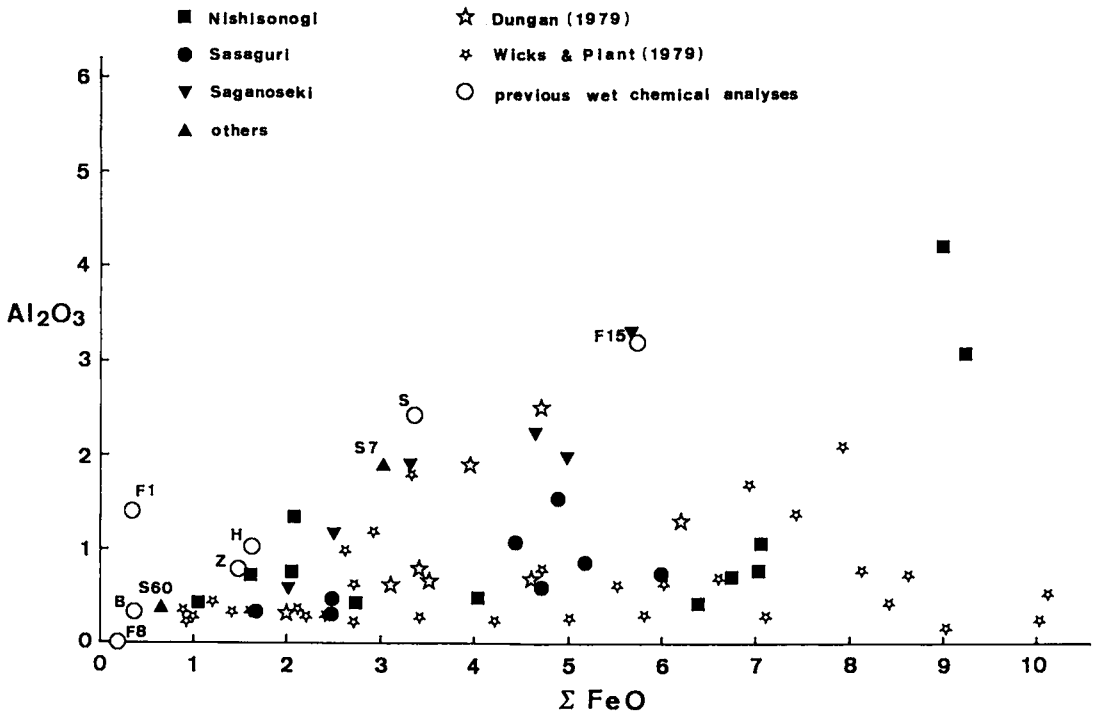
FIG. 6. Contents of Al_2O_3 versus ΣFeO (total iron expressed as Fe^{2+}) in antigorite from Saganoseki and in samples previously described in the literature. Nishisonogi, Sasaguri: Uehara & Shirozu (1985), F1, F8, F15: Faust & Fahey (1962), H: Hess *et al.* (1952), Z: Zussman (1954), B: Brindley & Santos (1971), S: Shimoda (1967).

Faust & Fahey 1962, Shimoda 1967, Brindley & Santos 1971) and by electron-microprobe analysis (Dungan 1979, Wicks & Plant 1979, Uehara & Shirozu 1985). Almost all of the antigorite samples contain less than 3 wt% Al_2O_3, whereas the range of ΣFeO contents varies from 1% to 10%. The ranges of ΣFeO contents in antigorite from Sasaguri, Nishisonogi, and Saganoseki are 1–6 wt%, 1–9 wt%, and 2–6 wt%, respectively.

The Al_2O_3 and ΣFeO contents of the Saganoseki antigorite are within the range of variation of the previous reported samples, but the Saganoseki antigorite tends to have relatively high $Al_2O_3/\Sigma FeO$ values. This could be a significant chemical factor associated with the formation of large supercells in the Saganoseki antigorite. Antigorite samples with a ΣFeO content in the range from 3 to 5 wt% have M in the range 8.8–9.6 in the Saganoseki samples (SG1, SG26B, SG48A), $M = 8.1$ in the Nishisonogi (N51B) and M in the range 6.9–7.8 (A59, A3, A103) in the Sasaguri samples. Further, the three Saganoseki samples are richer in Al content than the others.

Samples of lizardite–bastite after pyroxene have distinctly higher Al and Cr contents (Wicks & Plant 1979, Dungan 1979, Uehara & Naka 1983). Antigorite samples with a higher Al content (SG49 from Saganoseki, N81 and N1A from Nishisonogi) are also richer in Cr; therefore, these three antigorite samples may have been formed from relict Al- and Cr-rich lizardite–bastite.

Formation of large supercell

Kunze (1961) and Trommsdorff (1983) predicted that the shortening of the supercell parameter A (wavelength) of antigorite is connected with increasing metamorphic grade. Recently, Mellini *et al.* (1987) studied antigorite occurring in rocks formed under various metamorphic conditions and demonstrated the shortening of the supercell periodicities with metamorphic grade.

Uehara & Shirozu (1985) stated that samples with similar Fe^{2+} contents from Sasaguri and Nishisonogi have different parameters of the supercell and

observed that higher pressures and lower temperatures produced longer superstructures in specimens with the same Fe^{2+} content. When investigating the superstructure of antigorite, it is necessary to consider supercell parameters along with their chemical compositions.

The supercell parameter (M) of Fe-poor antigorite (ΣFeO in the range 1–3 wt%) is about 9.8 in the Saganoseki samples, 8.6 in samples from Nishisonogi and 7.8 in the Sasaguri antigorites. The metamorphic grade of the above three regions corresponds to the pumpellyite–actinolite facies, the glaucophane schist facies, and the epidote-amphibolite facies, respectively. The temperature of formation of the antigorite at Saganoseki is lower than that of two other localities. The condition of low temperature is probably also an important factor associated with the large superstructure, together with the $Al_2O_3/\Sigma FeO$ ratio.

Antigorite serpentinites occur widely across the low-grade to high-grade metamorphic regions. A phase diagram for serpentine reactions, taking account of field relations and available experimental results, has been constructed by Evans et al. (1976). Furthermore, an important reaction involving the calcium-bearing phases tremolite and diopside has been documented by Trommsdorff & Evans (1972), Oterdoom (1978) and Trommsdorff (1983). A lower temperature of formation of antigorite, by the reaction is tremolite + chrysotile → diopside + antigorite, has been found in Ca-bearing serpentinite, such as serpentinized lherzolite. Oterdoom (1978) estimated that the reaction takes place at 2 kbar and 192°C. The existence of tremolite in serpentinite at Saganoseki would reduce the temperature of formation of antigorite and increase the M value.

The samples SG1 and SG48A contain bastite and have large supercell parameters, but bastite-free samples SG6, SG21B and SG26B also have large supercell parameters. Recently, Chernosky et al. (1988) have shown by thermodynamic calculations that the reaction temperature of chrysotile → antigorite + brucite is displaced downward by about 50°C relative to the position calculated by Evans et al. (1976). The reaction takes place at 2 kbar and 250°C. The above two reactions probably are relevant to the formation of antigorite at Saganoseki.

The mineral assemblage of the Saganoseki serpentinites is mainly divided into two types, lizardite – chrysotile – antigorite with a small supercell parameter, and antigorite with a large supercell parameter. This could indicate that the reaction of large-supercell antigorite → small-supercell antigorite + chrysotile is coupled with the above two reactions.

The supercell parameters of the antigorite vary not only between different samples collected from a region in which the antigorite grains were formed at the same grade of metamorphism, but also within individual samples. The regional variation in M, mainly associated with variations in the content of Fe or Al (or both) in antigorite, is within ±1 within a region, but the M values of Fe-poor antigorite vary extensively from 9.0 to 7.0 according to metamorphic grade.

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