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Sadanagaite and magnesio-sadanagaite, new silica-poor members of calcic amphibole from Japan

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Abstract

Sadanagaite, $(K,Na)Ca_2(Fe^{2+},Mg,Al,Fe^{3+},Ti)_5[(Si,Al)_8O_{22}(OH)_2]$ where $Fe^{2+} \geq Mg$, $Al \geq Fe^{3+}$ and $Si < 5.5$, and its Mg-rich analogue, magnesio-sadanagaite $Fe^{2+} < Mg$, are extremely SiO_2-poor new amphiboles, which require the compositional extension of the edenite-pargasite series, and amending of the classification and nomenclature of amphiboles (Leake, 1978).

These are monoclinic, $C2$, Cm or $C2/m$; $a = 9.922(10)$, $b = 18.03(2)$, $c = 5.352(9)$Å, $\beta = 105.30(10)°$, $Z = 2$ for sadanagaite with $Si \simeq 5.0$, and $a = 9.964(2)$, $b = 18.008(3)$, $c = 5.354(2)$Å, $\beta = 105.55(2)°$, $Z = 2$ for magnesio-sadanagaite with $Si \simeq 5.0$. The strongest lines in the X-ray powder pattern for magnesio-sadanagaite are: 8.48(80)$(11\underline{0})$, 3.39(40)(131), 3.28(100)(240), 3.15(70)(310), 2.951(50)(221), 2.823(30)(330), 2.766(45)$(33\bar{1})$, 2.707(60)$(15\bar{1})$, 2.594(35)(061), 2.162(55)(261), 1.654(30)(461), these being similar to those of X-ray powder patterns of SiO_2-poor calcic amphiboles, especially kaersutite.

Both species have very similar physical properties, and are dark brown to black with a vitreous luster. Streak very light brown. Cleavage {110}, perfect. H (Mohs) ≃ 6. Density (calc.) 3.30 g/cm^3 for sadanagaite and 3.27 g/cm^3 for magnesio-sadanagaite. Optically biaxial positive with $2V = 80–90°$ (meas.), 86° (calc.) for sadanagaite, and with $2V$ close to 90° (meas.), 88° (calc.) for magnesio-sadanagaite. Refractive indices: $\alpha = 1.673(2)$, $\beta = 1.684(2)$, $\gamma = 1.697(2)$; $c{:}Z = 28°$ (sadanagaite), and $\alpha = 1.674(2)$, $\beta = 1.686(2)$, $\gamma = 1.699(2)$; $c{:}Z = 26°$ (magnesio-sadanagaite). Pleochroism strong with X' = pale brown to Z' = greenish brown (sadanagaite) and with X' = colorless to Z' = brownish yellow (magnesio-sadanagaite).

The minerals are found in two separate localities, Yuge and Myojin Islands, Japan, where they occur in Al-, Ti- and Fe-rich skarns in recrystallized limestone beds, forming bands with vesuvianite, spinel-hercynite, sphene, ilmenite and apatite in the former locality, and forming small lenses with titaniferous fassaite, vesuvianite, spinel-hercynite, sphene, ilmenite, magnetite, apatite and perovskite in the latter. Among them the peculiar composition of titaniferous fassaite is discussed with special reference to its extremely SiO_2-poor, Al_2O_3- and TiO_2-rich composition. The names are for Professor Ryoichi Sadanaga, Mineralogical Institute, University of Tokyo, in recognition of his outstanding contributions to the fields of mineralogy and crystallography.

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Introduction

The classification and nomenclature of amphiboles by Leake (1978) defines no lower limit of Si content in calcic amphiboles, although a prefix "subsilicic" is recommended when the number of Si is less than 5.75 on the basis of 23 oxygens. The occurrence of amphibole with Si = 5.27 has been reported by Appleyard (1975) and the necessity of new end member $NaCa_2Mg_3Al_2Si_5Al_3O_{22}(OH)_2$ ("Al-pargasite") has been suggested (Bunch and Okrusch, 1973; Doolan, Zen and Bence, 1978).

This contribution provides natural examples of amphiboles with lower Si than the above composition from two separate localities, Yuge and Myojin Islands, Ehime Prefecture, Japan. The composition of these amphiboles is so different from that of pargasite with regard to the number of Si that they are proposed as new Si-poor members (Si < 5.5) of the pargasite series. The minerals and mineral names, sadanagaite and magnesio-sadanagaite, have been approved by the Commission on New Minerals and Mineral Names, IMA (Commission Nos. 80-27 and 82-102). The names are for Emeritus Professor Ryoichi Sadanaga, Mineralogical Institute, University of Tokyo, in recognition of his outstanding contributions to the field of mineralogy and crystallography. Type specimens have been deposited in the Department of Mineralogy, the University Museum, University of Tokyo, Japan.

Both Yuge and Myojin are small islands located in the Seto Inland Sea, southwestern Japan (Fig. 1). In Yuge Island, where a limestone quarry has been operated, a sadanagaite-bearing vesuvianite skarn occurs as dark brown seams intercalated with limestone in the quarry. Myojin Island, one of islands known as the Shisaka Islands, consists largely of somewhat gneissose granitic rocks and gneiss with some limestone beds. Sato (1926) first reported the occurrence of clinopyroxene from skarn lenses in one limestone bed, and subsequently Kuno (1960) described this as titaniferous pyroxene, and Miyahisa et al. (1980) as fassaite. The dark colored lenses in the limestone bed include locally amphiboles of pargasite to magnesio-sadanagaite composition in addition to the above clinopyroxene.

Occurrence and associated minerals

According to the geological map compiled for Ehime Prefecture (1962), Yuge and Myojin Islands are situated near the boundary between the Paleozoic–Mesozoic Chichibu Belt and the Ryoke Metamorphic Belt (Fig. 1). Yuge Island belongs to the former belt with the intrusion of the Mesozoic Hiroshima-type granitic rocks. Myojin Island belongs to the latter belt, and weakly gneissose granitic rocks are associated with high temperature–low pressure-type metamorphic rocks. Both localities are only about 15 km from each other, and are thought to have similar geologic environments, although they are located in different geologic belts. No tectonic zone separates them, and the boundary between the belts is based only on the metamorphic grade.

In the Yuge limestone quarry, sadanagaite-bearing skarn occurs as seams in limestone with variable widths up to a maximum of 50 cm. The skarn seams usually have a symmetrical structure, that is, they consist of a wollas-

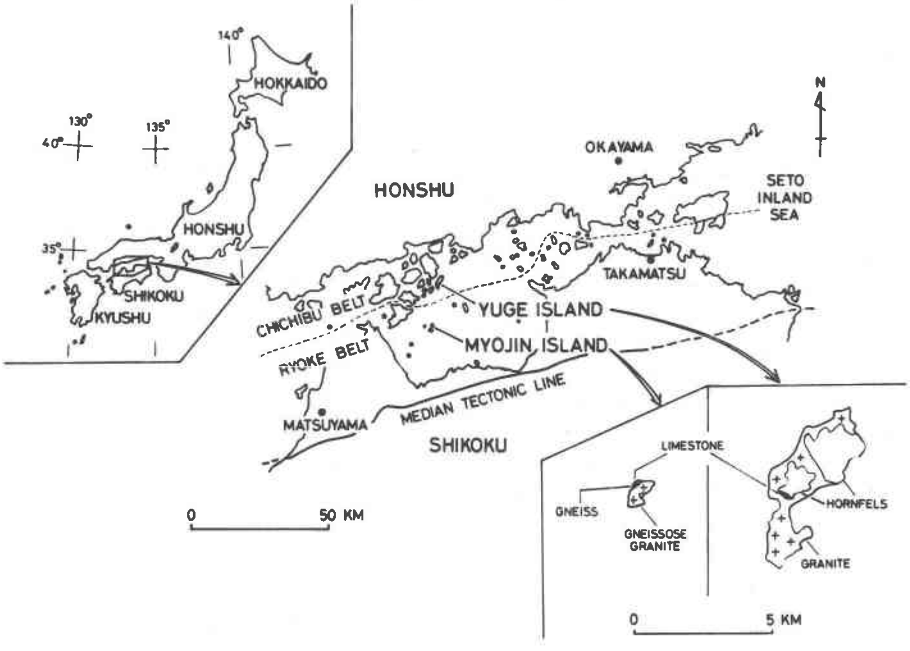

Fig. 1. Location and simplified geology of the Yuge and Myojin Islands, Ehime Prefecture, Japan.

tonite zone, a garnet–vesuvianite zone, and a vesuvianite–sadanagaite zone, traversing from limestone to the center of the seam. Sadanagaite may be lacking in the central vesuvianite zone of some seams. The sadanagaite-bearing zone occurs as a compact, dark brown band, consisting of vesuvianite and sadanagaite with minor amounts of spinel, sphene, ilmenite and apatite. Sadanagaite appears as isolated prismatic crystals with a maximum length of 1 mm or as polycrystalline aggregates in matrix vesuvianite. The composition of sadanagaites is quite variable with respect to SiO_2 as will be described in the following section and shown in Table 1.

Preliminary electron microprobe analyses show that the vesuvianite is rich in titanium with TiO_2 contents of 5 wt.% or more. The spinel is hercynite with variable amounts of the spinel molecule. Amounts of up to 5 wt.% Al_2O_3 are present in the sphene. Ilmenite has a small amount of the geikielite molecule.

In Myojin Island, the magnesio-sadanagaite-bearing skarn occurs as a lens or as an irregularly shaped nodule in crystalline limestone. Some lenses show a structure similar to that of boudinage. The size of the lens or nodule although variable ranges up to several tens of centimeters in width. The occurrence of the skarn is reported in detail as a fassaite-spinel rock by Miyahisa et al. (1980). As noted in their paper, clinopyroxene is the main constituent of the skarn, in contrast with the Yuge occurrence. The skarn includes, besides clinopyroxene, various amounts of spinel, vesuvianite, magnesio-sadanagaite to pargasite, phlogopite and minor amounts of sphene, ilmenite, magnetite, pyrrhotite, apatite and perovskite. Both clinopyroxene and magnesio-sadanagaite from this locality are short-prismatic to granular in shape with a maximum diameter of a few hundred microns.

The composition of the clinopyroxene has been debated since the first description by Sato (1926). Kuno (1960) showed that the clinopyroxene is rich in Al_2O_3 (6.89 wt.%) and TiO_2 (2.47%) by wet chemical analysis. Miyahisa et al. (1980) concluded that it is fassaite with up to 5 wt.% Al_2O_3 using the electron probe microanalyzer. Most of the skarn lenses contain clinopyroxenes with compositions similar to those given by Kuno (1960) and Miyahisa et al. (1980). The associated amphiboles are (ferroan) pargasite to less commonly magnesio-hastingsite.

The bulk chemical compositions of the lenses are quite variable, and in some rare lenses the compositions of the clinopyroxene and amphibole become very Si-poor, and Al- and Ti-rich. Magnesio-sadanagaite given in Table 1, No. 4, is an example from an extremely Si-poor lens. The associated clinopyroxene is also unusually Si-poor, with the number of Si = 1.3 on the basis of 6 oxygens. An

Table 1. Electron microprobe analyses of sadanagaites from the Yuge (Y) and Myojin (M) skarns

No.	1(Y)		2(Y)		3(Y)		4(M)		5(Y)		6(Y)	
SiO_2	30.2		29.9		31.1		32.1		33.5		29.5	
TiO_2	4.0		4.3		4.5		3.2		2.1		4.0	
Al_2O_3	24.8		22.6		22.5		22.0		19.7		23.2	
FeO*	15.2		17.4		16.2		13.7		17.8		16.4	
MnO	0.1		0.3		0.4		0.1		0.5		0.3	
MgO	6.6		6.1		6.6		8.0		6.4		7.7	
CaO	12.3		11.9		12.0		12.5		11.9		11.9	
Na_2O	1.0		0.6		0.6		0.7		1.2		0.7	
K_2O	3.7		3.7		3.6		3.8		3.1		3.5	
Total	97.9		96.8		97.5		96.1		96.2		97.2	
Numbers of cations on the basis of 23 oxygens												
Si	4.61	8.00	4.66	8.00	4.77	8.00	4.95	8.00	5.24	8.00	4.52	8.00
Al^{IV}	3.39		3.34		3.23		3.05		2.76		3.48	
Al^{VI}	1.07	5.00	0.81	5.02	0.85	5.01	0.94	4.93	0.88	5.00	0.71	5.06
Ti	0.46		0.50		0.51		0.38		0.25		0.46	
Fe^{3+}	0.40		0.63		0.48		0.39		0.39		0.97	
Fe^{2+}	1.55		1.63		1.60		1.37		1.94		1.13	
Mn	0.02		0.04		0.05		0.01		0.06		0.04	
Mg	1.50		1.41		1.52		1.84		1.48		1.75	
Ca	2.01	2.01	1.99	1.99	1.97	1.97	2.07	2.07	2.00	2.00	1.96	1.96
Na	0.29	1.00	0.17	0.91	0.19	0.88	0.22	0.97	0.37	0.99	0.20	0.88
K	0.71		0.74		0.69		0.75		0.62		0.68	

*Total iron. Nos. 1-3, 5: sadanagaite, 4: magnesio-sadanagaite, 6: Si-poor derivative of edenite-hastingsite series ($Al^{VI} < Fe^{3+}$).

Table 2. Electron microprobe analysis of titaniferous fassaite from the Myojin skarn, with cell dimensions and some optical properties

Oxide	wt. %	Numbers of cations on the basis of six oxygens		
SiO_2	34.0	Si	1.31	2.00
TiO_2	6.8	Al^{IV}	0.69	
Al_2O_3	18.6	Al^{VI}	0.16	0.98
FeO*	8.4	Ti	0.20	
MgO	6.1	Fe^{3+}	0.13	
CaO	25.0	Fe^{2+}	0.14	
Total	98.9	Mg	0.35	
		Ca	1.03	1.03

a=9.740(2) Å, b=8.829(2), c=5.311(1)
β=105.82°(1) based on 4-circle diffractometer data.
$2V_z \simeq 0°$, c:$Z \simeq 35°$, X': reddish purple,
Z': colorless to light yellow.
Dispersion : very strong.

* Total iron

electron probe microanalysis of such a clinopyroxene is given in Table 2. The amounts of MnO, Na_2O and K_2O are below the detection limit (<0.1 wt.%). Its composition is somewhat variable with Si ranging from 1.3 to 1.4 in one polished thin section. The amount of Fe^{3+} was estimated by calculation in order to satisfy the following two restrictions: O = 6 and $Al^{IV} = Al^{VI} + Fe^{3+} + 2Ti$. This unusually Si-poor fassaite is comparable to those reported by Dixon and Kennedy (1933) from a sphene-rich plagioclase–diopside hornfels xenolith in a norite, by Dowty and Clark (1973) from the Allende meteorite, and by Gross (1977) from an anorthite–wollastonite–gehlenite–pyroxene hornfels formed by thermal metamorphism of marls. The pyroxene in the Allende meteorite was formed under an extremely reducing environment, and includes a large amount of Ti^{3+}. The occurrence of fassaite in this study with about 1.3 Si confirms that one third of the Si can be replaced by Al^{IV} in the crystal structure of Ca-clinopyroxene under terrestrial Si-poor, and Al- and Ti-rich environments.

Not only the clinopyroxene but some other minerals associated with magnesio-sadanagaite in the Myojin skarn have unusually Al- and/or Ti-rich compositions, similar to those of the Yuge occurrence. Spinel from the Myojin skarn contains small amounts, up to 1 wt.% Cr_2O_3, in solid solution. Ilmenite usually includes several mole percent geikielite. Perovskite is associated with magnesio-sadanagaite in the Myojin skarn.

Chemistry

Because it was difficult to separate enough sadanagaites for conventional wet-chemical analysis, polished thin sections of sadanagaite-bearing skarns from both localities were prepared for electron probe analyses. X-ray intensities for Si, Ti, Al, Fe, Mn, Mg, Ca, Na and K were measured on standards and samples. The ratios were converted into oxide weight percentages using the method of Bence and Albee (1968). Detailed descriptions of the analytical procedure and the correction factors used have been given by Nakamura and Kushiro (1970). In the present study, the totals of the corrected weight percentages of oxides are usually in the range of 96.0 to 98.0%, with the remainder mainly due to H_2O. Qualitative checks showed some amount of fluorine but no appreciable amount of chlorine in these amphiboles.

The compositions of amphiboles from both localities range from sadanagaite to pargasite and less commonly to hastingsite. Common features among these amphiboles are: (1) complete occupancy of the B site by two calcium atoms, indicating that they are essentially calcic amphiboles, and (2) the predominance of potassium over sodium in the A site. Although the electron probe analyses lack ferrous and ferric iron determinations, meaningful estimation of the exact compositions was made by iterative calculations to satisfy the following two conditions: (1) total number of oxygens is 23.00, and (2) some Fe is taken as ferric so that $Al^{IV} = Al^{VI} + Fe^{3+} + 2Ti + Na + K$. Through such calculations, the number of cations in the C site usually comes close to 5.00, indicating the validity of this procedure.

Selected analyses (Table 1) demonstrate a wide range of Si from 5.5 to 4.5, and establish the definition of sadanagaite as a calcic amphibole ($(Ca + Na) \geq 1.34$, $Na_B < 0.67$) with $Si < 5.5$, $(Na + K)_A \geq 0.5$, $Al^{VI} \geq Fe^{3+}$ and $Ti < 0.50$ per unit cell formula. This requires an extension to the silica-poor side of the edenite-pargasite series (after Leake, 1978) as shown in Figure 2. Sadanagaites with $Fe^{2+} \geq Mg$ are common in the Yuge skarns, and those with $Fe^{2+} < Mg$, for which the name magnesio-sadanagaite is given, are typically found in the Myojin occurrence (Table 1).

In some analyses, such as No. 6 in Table 1, the relation $Al^{VI} < Fe^{3+}$ is obtained after the calculation of the estimated amount of ferric iron. According to Leake's (1978) nomenclature, such amphiboles with $Al^{VI} < Fe^{3+}$ could be a new species corresponding to the Si-poor derivative of the edenite-hastingsite series. Such amphiboles, in the present study, are rather rare, and the calculated excess of Fe^{3+} over Al^{VI} is not significant if we consider the error included in the estimation of the amount of ferric iron. The establishment of new species at the silica-poor side of the edenite–hastingsite series must remain for future studies. Although sadanagaites are defined as having $Ti < 0.5$ in the formula, analyses show that the number of Ti may exceed 0.5 (e.g. Nos. 2 and 3 in Table 1). According to Leake's (1978) nomenclature, such analyses represent Si-poor derivatives of kaersutite. The excess of Ti over 0.5 is very small in these samples, and we will not discuss the status of Si-poor derivatives of kaersutite.

In sadanagaites from both localities, the amount of K

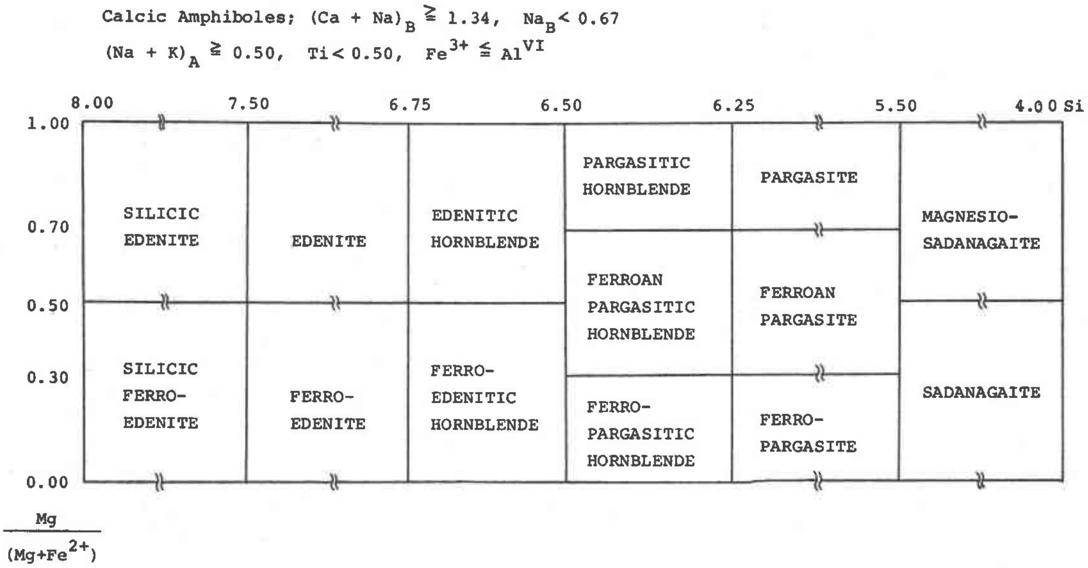

Fig. 2. The position of sadanagaite and magnesio-sadanagaite in the classification of calcic amphiboles after Leake (1978).

always exceeds that of Na. According to Leake's (1978) suggestion, potassium ($K \geq 0.50$) or rarely potassian ($K = 0.25$–0.49) (magnesio-)sadanagaite is the recommended name for the present amphiboles. In experimental investigation of the Na–K substitution in some amphiboles Hinrichsen and Schürmann (1977) failed to synthesize pargasite with $Na_{50}K_{50}$ to Na_0K_{100} at temperatures between 750–1000°C and pressures between 1 and 4 kbar. The authors, however, suggested that extensive substitution of Na by K in pargasitic amphiboles would not be impossible, as shown in the present study.

Because all sadanagaites in this study have high K/Na ratios, large replacement of Si by Al in the T site may be crystallochemically necessary for the entry of K into the A site. It is uncertain whether high K substitution in the A site is an essential feature of the Si-poor member of calcic amphiboles. Appleyard (1975), however, gives an example of Na-rich amphibole in the hastingsite series with $Si = 5.27$ from metasomatic alkaline gneisses at Wolfe, Ontario. Because the significance of the K/Na ratio in the amphibole structure has not yet been established, the large K/Na value should not be included in the definition of sadanagaite.

As noted above, amphiboles in the Yuge skarn are chiefly associated with vesuvianite. Their Si content ranges from 4.5 to 6.0, that is, from sadanagaite to ferroan pargasite. Extremely low Si contents are caused partly by the relatively high Ti contents in the mineral (e.g. Nos. 1–3 and 6 in Table 1). On the other hand, amphiboles in the Myojin skarn are usually associated with clinopyroxenes, and Ti tends to be concentrated in clinopyroxene rather than the associated amphibole. Analysis No. 4 in Table 1 is an example of an amphibole poorest in Si (≃5.0) from the Myojin skarn. This amphibole is associated with the titaniferous fassaite, the composition of which is given in Table 2. The amphiboles from the Myojin skarn tend to be Mg-rich, and range mainly from magnesio-sadanagaite to pargasite.

Table 3. X-ray powder diffraction data for magnesio-sadanagaite

I_o	d_o Å	d_c Å	hkl	I_o	d_o Å	d_c Å	hkl
10	9.00	9.00*	020	10	1.909	1.909*	510
80	8.48	8.47*	110	3	1.892	1.893*	$46\bar{1}$
5	4.97	4.96*	$11\bar{1}$	3	1.865	1.867	$44\bar{2}$
15	4.80	4.80*	200			1.865	$19\bar{1}$
10	4.50	4.50*	040	5	1.829	1.829*	530
3	4.01	4.01*	111	5	1.801	1.801*	0.10.0
40	3.39	3.39*	131	2	1.778	1.778*	$51\bar{2}$
100	3.28	3.28*	240	2	1.715_B	1.713	332
70	3.15	3.15*	310			1.711	$31\bar{3}$
50	2.951	2.949*	221	5	1.695_B	1.696	$28\bar{2}$
30	2.823	2.823*	330	2	1.685	1.684*	$39\bar{1}$
45	2.766	2.768*	$33\bar{1}$	30	1.654	1.654*	461
60	2.707	2.708*	151	5	1.643	1.642*	480
35	2.594	2.594*	061	3	1.614	1.614*	1.11.0
15	2.578	2.578*	$20\bar{2}$	5	1.599	1.600	600
13	2.392	2.392*	350			1.594	$15\bar{3}$
16	2.355	2.355	$42\bar{1}$	10	1.559	1.560	402
		2.358	$35\bar{1}$			1.560	$60\bar{2}$
5	2.317_B	2.318	$31\bar{2}$	3	1.529	1.529*	$26\bar{3}$
3	2.238_B	2.237	$24\bar{2}$	6	1.507	1.507*	551
55	2.162	2.163*	261	3	1.476_B	1.476	153
12	2.051	2.054	202			1.476	$2.10.\bar{2}$
		2.052	$40\bar{2}$	5	1.457	1.457*	3.11.0
15	2.020	2.021*	351	10	1.452	1.452*	$66\bar{1}$
5	2.004	2.005*	370	5	1.370	1.370*	512

* Included in the least squares refinement of the unit cell dimensions

X-ray crystallography and physical properties

X-ray single crystal studies showed these amphiboles to be monoclinic, with possible space group *C*2, *Cm* or *C*2/*m*. An X-ray powder diffraction diagram of magnesio-sadanagaite was recorded with a powder diffractometer, whereas the pattern of sadanagaite was obtained with a Gandolfi camera due to the very small quantity of available material. Least squares refinements of the unit cell dimensions from about 20 measured reflections for sadanagaite and from over 30 for magnesio-sadanagaite gave the following values: a = 9.922(10), b = 18.03(2), c = 5.352(9)Å, β = 105.30(10)° for sadanagaite; and a = 9.964(2), b = 18.008(3), c = 5.354(2)Å, β = 105.55(2)° for magnesio-sadanagaite. No essential difference in the powder pattern is recognized between magnesio-sadanagaite (Table 3) and sadanagaite; their patterns are also similar to those of Si-poor calcic amphiboles such as kaersutite given in JCPDS Card No. 17-478. The calculated density is approximately 3.30 (g/cm^3) for sadanagaite and 3.27 (g/cm^3) for magnesio-sadanagaite.

Both species have very similar physical properties, dark brown to black in color with a vitreous luster. The color of the streak is very light brown. Cleavage {110} is perfect, and characteristic amphibole cleavages are obvious in thin sections. The Mohs hardness is approximately 6. Sadanagaite as well as magnesio-sadanagaite is optically biaxial; positive with $2V$ = 80–90° (obs.), 86° (calc.) for sadanagaite, and with $2V$ close to 90° (obs.), 88° (calc.) for magnesio-sadanagaite. The refractive indices are α = 1.673(2), β = 1.684(2), γ = 1.697(2), c: Z = 28° (sadanagaite) and α = 1.674(2), β = 1.686(2), γ = 1.699(2), c:Z = 26° (magnesio-sadanagaite). Both are pleochroic under the microscope from X' = pale brown to Z' = greenish brown (sadanagaite) and from X' = colorless to Z' brownish yellow (magnesio-sadanagaite). The color under the microscope and pleochroism become paler and weaker, respectively, with increasing Si content of sadanagaites. These properties for sadanagaite were obtained using grains with some compositional variations. As sadanagaite with Si close to 5.0 is predominant in the specimen, it is concluded that these properties represent those for sadanagaite with Si $\simeq$ 5.0. The properties for magnesio-sadanagaite are thought to also represent those for magnesio-sadanagaite with Si $\simeq$ 5.0, since they have been measured using grains separated from the specimen which has been found to be relatively homogeneous and provided the chemical composition of analysis no. 4 in Table 1.

Genetic considerations

Miyahisa et al. (1980) analyzed the compositions of magnetite and ilmenite associated with fassaite from the Myojin locality. On the basis of Buddington and Lindsley's (1964) geothermometer, they estimated the temperature of formation to have been 600°C, and the oxygen fugacity somewhat greater than 10^{-20} atm, reflecting a rather reducing environment. On the basis of a bulk chemical analysis of the skarn lens, they concluded that the amphibolite facies rock was originally derived from an Al-rich sediment such as a laterite, intercalated with limestone. The similar nature of the Yuge skarn strongly suggests that it was also derived from similar Al–Fe–Ti-rich sediments intercalated with limestone. Hydrothermal solutions may have aided the exchange of material between the aluminous sediments and the surrounding limestone. According to Iwao (1978), occurrences of Fe–Al–Ti-rich metamorphic rocks are known widely in the late Paleozoic to Triassic formations of Japan. The Yuge and Myojin occurrences are additional examples of lateritic sediments formed during these times.

Acknowledgments

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