Bull. Natl. Mus. Nat. Sci., Ser. C, 41, pp. 45–52, December 22, 2015

An okenite-like mineral from Anijima Island, Ogasawara Islands, Tokyo, Japan

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Abstract An okenite-like mineral occurs in amygdules in hyaloclastite corresponding to boninite component at the Uguisu Beach, Anijima Island, Ogasawara Islands, Tokyo, Japan. It is found as an aggregate of pale pinkish yellow to white fibrous crystals in association with apophyllite. The representative chemical analysis excluding H_2O gave SiO_2 57.14, Al_2O_3 0.19, CaO 24.56, MgO 0.62, Na_2O 1.83, K_2O 0.82, F 0.42, $-O=F_2$ 0.18, total 85.40 wt.%. The empirical formula is; $(Ca_{4.13}Na_{0.56}K_{0.17}Mg_{0.14})_{\Sigma 5.00}(Si_{8.97}Al_{0.03})_{\Sigma 9}[O_{22.52}F_{0.21}]_{\Sigma 22.73}$ on the basis of $Si+Al=9$. The unit cell parameters calculated from the powder X-ray diffraction peaks are; $a=9.7381(14)$, $b=7.2870(9)$, $c=19.209(2)$ Å, $\alpha=90.777(11)$, $\beta=100.824(11)$, $\gamma=111.438(11)°$, $V=1241.2(3)$ Å^3. We proposed a new crystal structure for the okenite-like mineral different with that of the known okenite.

Key words: an okenite-like mineral, hyaloclastite, Anijima, Ogasawara

Introduction

In October, 2013 we collected an okenite-like mineral, apophyllite and some zeolites in hyaloclastite at the Anijima Island, Ogasawara Islands, during the mineralogical survey as a part of the project on the Geologic Research and Sufficiency of National Rock and Mineral Samples in Ogasawara Arc. This research accessed budgetary support from Director General, National Museum of Nature and Science.

Okenite is not so rare and occurs generally in amygdules in basaltic rocks. The finest specimens are ball-like aggregates found in amygdules in the Deccan basalt from around Poona, Maharashtra, India. Although Von Kobell named ockenit (later shortened to okenit or okenite) to the material from Disko Island, Greenland in 1828, refinement of the crystal structure for okenite from Kolhapur, Maharashtra, India was in 1983 by Merlino. In Japan the occurrences of okenite were reported from Nagasaki and Kita-hatsuneura, Chichijima Island, Ogasawara Islands (Nishido *et al.*, 1983), Nakazato, Aomori Prefecture (Aoki, 1984) and Shimo-Hatsukari, Yamanashi Prefecture (Kato, 1973; Aoki, 1984).

The present paper deals with the occurrence of an okenite-like mineral, and discusses on the structural model estimated from the X-ray diffraction pattern and the chemical compositions in comparison with typical okenite and the related calcium hydrous silicates such as gyrolite, truscottite and reyerite.

Occurrence

The Bonin Islands represent an emergent section of the Izu–Bonin fore-arc basement (Fig. 1A). They form parts of subaerial basement exposure extending from ca. 100 km south of the mainland of Japan, to the southern Ioto Island. This subaerial basement is separated by fore-arc of the Bonin Islands (Mukojima, Chichijima and

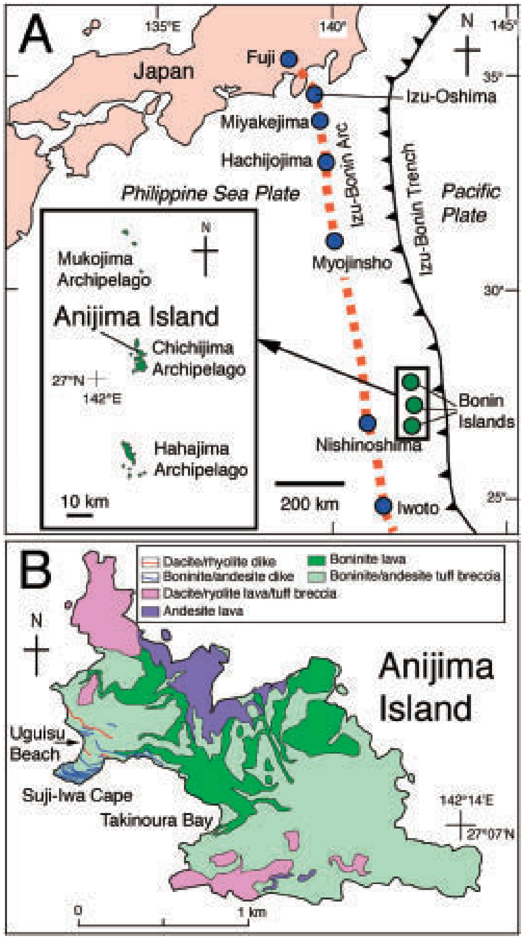

Fig. 1. (A) Map of the Izu–Bonin Islands. The inset is a map showing the location of Anijima Island in Bonin Islands. (B) Geological map of Anijima Island (after Umino and Nakano, 2007).

Fig. 2. Outcrop of sample locality.

Fig. 3. Fibrous aggregate of “okenite” is rimmed by celadonite in amygdule. “Okenite” aggregate is approximately 25 mm in width.

Fig. 4. “Okenite” aggregate fills cavity of apophyllite crystals in amygdule. Specimen is approximately 30 mm in width.

Hahajima archipelagos) and an active volcanic arc (Izu–Bonin Arc). Subaerial exposures on the Bonin Islands have been studied extensively, and are where boninites were described Chichijima and Mukojima archipelagos are large piles of boninites and slightly younger andesite and dacite (e.g., Kuroda and Shiraki, 1975).

Along the entire Izu–Bonin fore-arc, mid-ocean ridge basalt–like proto-arc basalt extruded at 52–48 million years ago (Ma), followed by arc tholeiitic lavas on the Bonin Islands (Umino and Nakano, 2007). The dissected tholeiitic rocks are overlain by 48–46 Ma highly depleted, high-silica boninite with minor low-silica boninite (Maruberiwan Formation), which became covered with less-depleted, low-silica boninite associated with calc-alkalic magmas at 45 Ma (Mikazukiyama Formation). By 44 Ma, the volcanic front had retreaded westward and fertile arc magmas on the Hahajima Archipelago had superseded boninites (Kanayama *et al.*, 2014).

The Anijima Island (27°7′30″N, 142°11′28″E), north of the Chichijima Island, is mainly composed of hyaloclastite (i.e., tuff breccia) and pillow lavas of the Mikazukiyama Formation (Fig. 1B). One important geological description on the Anijima Island is that several boninite and/or bronzite andesite dikes are present along the Suji-Iwa Cape and Uguisu Beach (Fig. 1B).

The Uguisu Beach is located at west coast of the Anijima Island, Ogasawara Islands about 1000 km south of central Tokyo metropolitan. At the beach are observed hyaloclastite and dikes formed by Paleogene volcanic activities.

In October, 2013 we visited the Uguisu Beach in order to research the occurrence of rocks and minerals. Here is characterized in brownish green enstatite (bronzite) grains as major constituents of beach sand. The beach looks brownish green in color that resembles feather color of a bird, Japanese bush warbler (Japanese name is uguisu). A lot of amygdules are observed in hyaloclastite at the cliff continued from north end of the beach (Fig. 2). We recognize the occurrence of apophyllite, heulandite, phillipsite, chalcedonic quartz and an okenite-like mineral there. We describe the okenite-like mineral as "okenite" as follows. The apophyllite is rarely found as colorless short square prisms less than 20 × 30 mm across. Also aggregates of fine prismatic crystals of apophyllite commonly occur as amygdule fillings.

The "okenite" was found as amygdule fillings composed of aggregate of very fine, fibrous crystals (Fig. 3). It is pinkish yellow to greyish white in color. The "okenite" also occurs on apophyllite crystals (both of apophyllite-(KF) and apophyllite-(KOH) were recognized) and fills in an open space of amygdules (Fig. 4). The amygdules are commonly surrounded by green-color, massive celadonite.

X-ray Crystallography

X-ray powder diffraction data of the "okenite" were obtained using a Gandolfi camera with a diameter of 114.6 mm and Ni-filtered Cu*Kα* radiation. The data were recorded on an imaging plate, and processed with a Fuji BAS-2500 bioimage analyser using a computer program written by Nakamuta (1999). The X-ray powder diffraction patterns were also measured by the Rigaku SmatLab powder diffarctometer in a Debye-Scherrer geometry using Cu*Kα*1 radiation from a primary monochromator. The sample was filled in a lithium borate (Lindemann glass) capillary tube with an inner diameter of 0.9 mm, and

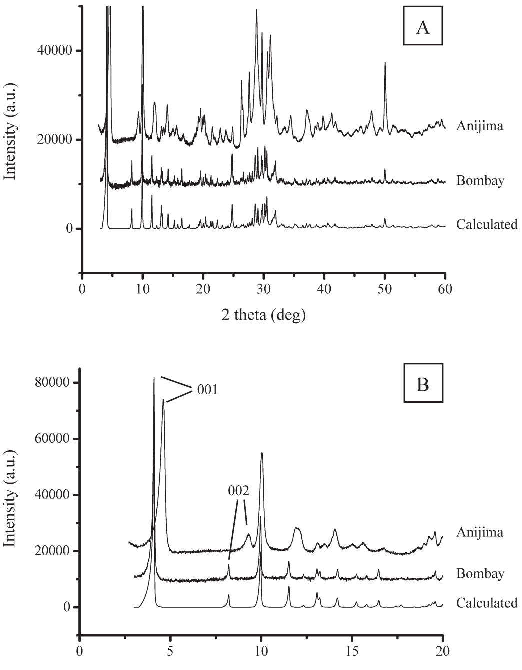

Fig. 5. X-ray powder diffraction pattern of "okenite" from the Anijima Island, a reference pattern of typical okenite from Bombay, India, and theoretical pattern calculated from structure parameters of okenite (Merlino, 1983). (A) 2θ range up to 60° and (B) enlarged view of low-angle region. Backgrounds of observed diffraction data were subtracted for better visibility.

Table 1. List of X-ray diffraction peaks of “okenite”.

d	I/I_0	*hkl*	*d*	I/I_0	*hkl*
19.19	100	001	3.228	33	105
9.49	12	002	3.094	59	$2\bar{2}2$, $21\bar{3}$, $02\bar{3}$, 022
8.80	66	$10\bar{1}$	3.005	49	$30\bar{2}$
7.43	16	101, $10\bar{2}$	2.916	41	$21\bar{4}$, $1\bar{1}\bar{6}$
6.76	6	010, $1\bar{1}0$	2.873	47	$2\bar{2}3$, $02\bar{4}$
6.57	5	$01\bar{1}$	2.826	17	301, $2\bar{2}\bar{4}$, $3\bar{2}\bar{1}$, 120, $11\bar{6}$, $12\bar{2}$
6.29	15	003	2.779	13	$3\bar{2}\bar{2}$, 106
5.89	6	102	2.683	8	$3\bar{2}\bar{3}$, 007, $3\bar{1}\bar{5}$
5.68	7	$10\bar{3}$, $1\bar{1}\bar{2}$	2.601	12	122, $3\bar{2}2$, $1\bar{2}5$, $30\bar{5}$
5.28	4	$1\bar{1}2$, 012	2.551	3	$1\bar{1}\bar{7}$, $3\bar{2}\bar{4}$
4.746	5	$1\bar{1}\bar{3}$	2.422	14	$12\bar{5}$, $4\bar{1}\bar{1}$, $2\bar{2}5$, 017
4.674	8	103, $11\bar{1}$	2.396	10	$31\bar{3}$, $2\bar{3}0$, $1\bar{3}1$, 310, $10\bar{8}$, $3\bar{2}\bar{5}$
4.607	12	$2\bar{1}\bar{1}$, 110	2.335	7	206, $2\bar{3}\bar{2}$, $22\bar{1}$, $31\bar{4}$
4.532	16	$20\bar{1}$	2.312	8	$4\bar{1}\bar{4}$, $4\bar{1}1$, $4\bar{2}\bar{1}$, $1\bar{3}\bar{2}$
4.434	13	$11\bar{2}$, 200	2.264	11	$1\bar{1}\bar{8}$, $40\bar{2}$, $4\bar{2}\bar{3}$, $3\bar{1}\bar{7}$, $12\bar{6}$, $03\bar{1}$
4.384	13	$2\bar{1}\bar{2}$, $20\bar{2}$	2.188	12	$4\bar{2}\bar{4}$, $40\bar{4}$
4.318	6	111, $2\bar{1}1$	2.156	9	018, $3\bar{3}\bar{3}$, $2\bar{3}\bar{4}$, 222
4.124	9	201	2.115	5	207, $40\bar{5}$, $1\bar{3}\bar{4}$
4.041	5	$11\bar{3}$, $20\bar{3}$, $01\bar{4}$	2.059	4	402, 033, $20\bar{9}$, $30\bar{8}$
3.897	9	$2\bar{1}2$, 112	1.998	6	$4\bar{2}\bar{6}$, $4\bar{3}\bar{2}$, $31\bar{7}$
3.743	9	005, $10\bar{5}$, $1\bar{1}4$	1.968	7	$3\bar{2}6$, 131, $3\bar{3}4$, $2\bar{1}8$, 403, $4\bar{3}\bar{3}$, $4\bar{3}1$
3.580	10	$1\bar{2}1$	1.935	8	$1\bar{5}\bar{2}$, $41\bar{3}$, $41\bar{1}$
3.382	30	$1\bar{2}\bar{2}$, $1\bar{1}\bar{5}$, $02\bar{1}$	1.900	14	$30\bar{9}$, $5\bar{2}\bar{3}$, 320, $5\bar{1}0$
3.357	19	$2\bar{2}0$, $2\bar{2}\bar{1}$	1.849	8	$01\overline{10}$, $5\bar{1}1$, $11\overline{10}$, $02\bar{9}$, 028, $5\bar{2}1$, $5\bar{1}\bar{5}$, $4\bar{3}3$, 133
3.255	14	$21\bar{1}$	1.821	35	$2\bar{4}0$, $1\bar{2}9$

a high-speed 1D silicon strip detector (D/tex Ultra 250) was used with continuous scan mode of 0.1° per minute and data intervals of 0.01°. The diffraction data were analyzed by the Rietveld refinement program RIETAN-FP (Izumi and Momma, 2007). Figure 5A shows the diffraction data measured by the Debye-Scherrer diffarctometer as well as that of a reference okenite sample from Bombay, India, and theoretical diffraction pattern of okenite calculated from structure parameters reported by Merlino (1983). The most notable difference between a diffraction pattern of the present sample and that of the reference okenite is positions of the lowest 001 and 002 reflections (Fig. 5B). The unit cell parameters refined by Le Bail analysis of the powder X-ray diffraction data are; $a = 9.7381(14)$, $b = 7.2870(9)$, $c = 19.209(2)$ Å, $\alpha = 90.777(11)$, $\beta = 100.824(11)$, $\gamma = 111.438(11)$°, $V = 1241.2(3)$ Å^3. Compared with the unit cell parameters of okenite, length of the *c*-axis of the present sample is noticeably short. List of X-ray diffraction peaks and their relative intensities is summarized in Table 1.

Chemical Composition

Chemical analysis of the “okenite” was carried out using an INCA Oxford energy dispersive X-ray spectrometer installed in JSM-6610 SEM. The standard materials used were wollastonite for Si, Al_2O_3 for Al, wollastonite for Ca, MgO for Mg, NaCl for Na, KBr for K and CaF_2 for F, respectively. No other elements were observed in the EPMA analysis. Table 2 shows results for five analytical points of the present “okenite” in comparison with okenite from another localities. The representative empirical formula (No. 3) of the “okenite” is $(Ca_{4.13}Na_{0.56}K_{0.17}Mg_{0.14})_{\Sigma5.00}(Si_{8.97}Al_{0.03})_{\Sigma9}[O_{22.52}F_{0.21}]_{\Sigma22.73}$ on the basis of Si + Al = 9 and excluding H_2O. If it is presumed that the difference from 100 wt.% in “okenite” (3 in Table 2) is H_2O, the empirical formulae is $(Ca_{4.13}Na_{0.56}K_{0.17}Mg_{0.14})_{\Sigma5.00}(Si_{8.97}Al_{0.03})_{\Sigma9}[O_{22.52}F_{0.21}]_{\Sigma22.73}\cdot7.64H_2O$. According to the crystal structure analysis by Merlino (1983), the ideal formula of okenite is demonstrated as $Ca_{10}Si_{18}O_{46}\cdot18H_2O$ $(Ca_5Si_9O_{23}\cdot9H_2O)$. The present “okenite”

Table 2. Chemical composition of “okenite” from the Anijima Island and okenite from Bombay, India (Gard and Taylor, 1956).

	1	2	3	4	5	6	7
SiO_2	55.79	58.08	57.14	57.26	56.24	53.88	55.00
Al_2O_3	0.15	0.23	0.19	0.22	0.22	0.08	
Fe_2O_3	0	0	0	0	0	0.01	
CaO	24.28	24.26	24.56	24.37	24.28	27.61	28.51
SrO	0	0	0	0	0	0.27	
MgO	0.63	0.58	0.62	0.60	0.71	0	
Na_2O	1.75	2.30	1.83	2.26	1.55	0.12	
K_2O	0.86	0.91	0.82	0.85	0.79	0.06	
F	0.55	0.50	0.42	0.53	0.54	0	
H_2O	nd	nd	nd	nd	nd	18.02	16.49
$-O=F_2$	0.23	0.21	0.18	0.22	0.23		
total	83.78	86.65	85.40	85.87	84.10	100.05	100.00
on the basis of (Si + Al) = 9							
Si	8.97	8.96	8.97	8.96	8.96	8.98	9
Al	0.03	0.04	0.03	0.04	0.04	0.02	
Fe						0	
Ca	4.19	4.01	4.13	4.08	4.15	4.93	5
Sr						0.03	
Mg	0.15	0.13	0.14	0.14	0.17		
Na	0.55	0.69	0.56	0.68	0.48	0.04	
K	0.18	0.18	0.17	0.17	0.16	0.01	
∑	5.07	5.01	5.00	5.07	4.96	5.01	
F	0.28	0.24	0.21	0.26	0.27		

1–5: Uguisu Beach, islet of Anijima, Ogasawara Islands, Japan
6: Bombay, India
7: $Ca_5Si_9O_{23}\cdot 9H_2O$

is characterized as alkali- and Al-bearing okenite, if amount of H_2O is disregarded.

The representative chemical compositions of apophyllite in association with the “okenite” are demonstrated in Table 3.

Table 3. Chemical composition of apophyllite associated with “okenite”.

	apophyllite	
	− (KF)	− (KOH)
SiO_2	50.05	50.97
Al_2O_3	0.05	0.09
CaO	24.17	24.42
Na_2O	0	0.19
K_2O	5.01	5.03
F	1.90	0.40
H_2O*	0.05	0.76
$-O=F_2$	0.80	0.17
total	80.43	81.69
Si	7.90	7.89
Al	0.01	0.02
Ca	4.08	4.05
Na		0.05
K	1.01	0.99
∑	13	13
F	0.95	0.20
H	0.05	0.80

*: Calculation on the basis of (F + OH)=1 and stoichometry excluding H_2O molecule.

Discussion

Although the X-ray powder diffraction pattern of the “okenite” can be indexed with unit cell parameters similar to okenite, there are substantial differences between positions and intensities of diffraction peaks of the present sample and those of typical okenite. Length of the *c*-axis of the present sample is about 2.8 Å shorter than previously reported values of okenite. We tried Rietveld analysis of the XRD data adopting structural model of okenite (Merlino, 1983) but could not yield fairly good fit ($R_{wp} = 8.28\%$, $S = 12.36$, $R_B = 19.28\%$, $R_F = 6.47\%$). The crystal structure of okenite is comprised of alternating Ca layers and SiO_4 layers parallel to the *a*–*b*

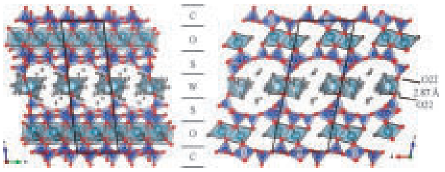

Fig. 6. Crystal structure of okenite viewed along the *a*- and *b*-axes drawn by VESTA (Momma and Izumi, 2011).

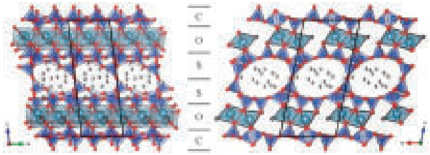

Fig. 7. A hypothetical structural model of “okenite” derived by removal of Ca-water layer of okenite.

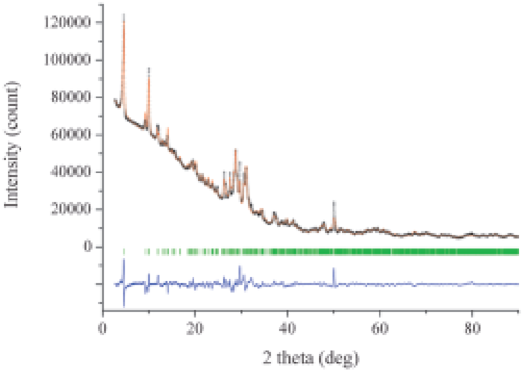

Fig. 8. Observed (cross marks), calculated (solid line), and difference patterns (solid line at the bottom) of the Rietveld refinement of “okenite”. The tick marks below the observe pattern represent the calculated peak positions.

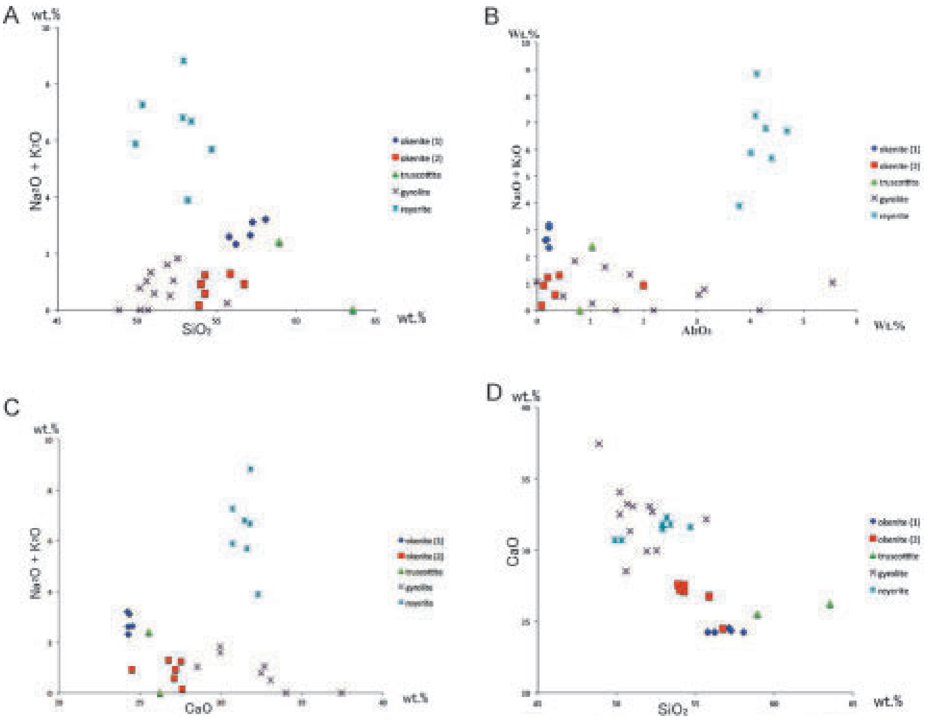

Fig. 9. (A) SiO_2 vs. $Na_2O + K_2O$ diagram in okenite (1), okenite (2), truscottite, gyrolite and reyerite. Data source: okenite (1) (present study), okenite (2) (Gard and Taylor, 1956; Sukheswala *et al.*, 1974; Nishido *et al.*, 1983; Aoki, 1984), truscottite (Hövig, 1914; Minato and Kato, 1967), gyrolite (Anderson, 1851; Macky and Taylor, 1953; Mizota, 1969; Kobayashi and Kawai, 1974; Sukheswala *et al.*, 1974; Noack, 1983; Merlino, 1988) and reyerite (Chalmers *et al.*, 1964; Clement and Ribbe, 1973; Livingstone, 1988). (B) Al_2O_3 vs. $Na_2O + K_2O$ diagram. Data sourse:ditto. (C) CaO vs. $Na_2O + K_2O$ diagram. Data sourse:ditto. (D) SiO_2 vs. CaO diagram. Data sourse:ditto.

plane (Fig. 6). There are two types of SiO_4 layers; one of them is a tetrahedral sheet (S), and the other is composed of three-repeat double chains (C) parallel to the *b*-axis. The CaO_6 octahedral double chain (O) is interconnecting the two types of SiO_4 layers, and the Ca-water layer (W) is connecting the adjacent two tetrahedral sheets. Then the periodicity of okenite along the *c*-axis can be expressed by using these four components as COSWSOC. A distance between two oxygen atoms (O22) connected by disordered Ca atoms in the Ca-water layer is 2.87 Å, which corresponds to the difference of length of the *c*-axis between the present sample and typical okenite. If the Ca-water layer is removed and the adjacent SiO_4 sheets are directly bridged by single O22 atom, zeolitic channels parallel to the *b*-axis are formed and the resulting structure has unit cell parameters that are very close to the observed values (Fig. 7). Then this structural model was also tested by Rietveld analysis. Atomic positions of S, C, and O layers in the new structural model were derived from structural model of okenite by converting fractional coordinates to keep 3D geometry. The positions and atomic displacement parameters of these atoms were fixed during the refinement. Because there is no information about positions of water molecules and other cations in the zeolitic channel, occupancies and positions of these atoms were refined by trial and error. The final fit of the refined model to the observed data ($R_{wp}=4.16\%$, $S=6.20$, $R_B=2.72\%$, $R_F=0.76\%$) was significantly better than the fit by structural model of okenite. This result suggests the proposed structural model as a fairly good approximation of the true structure of "okenite", although the reliability of the refinement is still not sufficient (Fig. 8), most probably due to poorly recovered guest atoms in the zeolitic channels and also due to possible distortion of tetrahedral and octahedral layers.

The occurrence and appearance of truscottite, gyrolite and reyerite resemble those of okenite, and it is often difficult to distinguish among them. Moreover the similarities of powder X-ray diffraction patterns and chemical compositions are pointed out.

Figures 9A, B, C and D indicate diagrams of SiO_2 vs. Na_2O+K_2O, Al_2O_3 vs. Na_2O+K_2O, CaO vs. Na_2O+K_2O and SiO_2 vs. CaO, respectively. In these figures okenite (1)(= "okenite") indicates specimens from the Anijima Island and those from other localities are shown as okenite (2). In Figure 9A the compositional range of the "okenite", okenite, truscottite, gyrolite and reyerite are distinguishable except one example of gyrolite (SiO_2 55.68, Na_2O+K_2O 0.24) from Mururoa, French Polynesia (Noack, 1983). In Fig. 9B the compositional range of the "okenite", okenite, truscottite and gyrolite are indistinct except reyerite. Al_2O_3 content of gyrolite varies widely, but it is plotted from 0.71 to 4.18 if neglecting one example (Al_2O_3 0, Na_2O+K_2O 1.05) from Bombay, India (Sukheswala *et al.*, 1974). Contrary to gyrolite, Al_2O_3 content of the "okenite" and okenite do not vary if neglecting one example (Al_2O_3 2.00, Na_2O+K_2O 0.92) from Kita-hatsuneura, Chichijima Island (Nishido *et al.*, 1983). In CaO vs. Na_2O+K_2O diagram (Fig. 9C) the compositional ranges of the "okenite", okenite and truscottite are indistinct, but those of gyrolite and reyerite are distinct. There is, however, reverse relations among them in SiO_2 vs. CaO (Fig. 9D). Although the compositional range among the "okenite", okenite, truscottite, gyrolite and reyerite are, on the whole, distinct, the more detailed con-

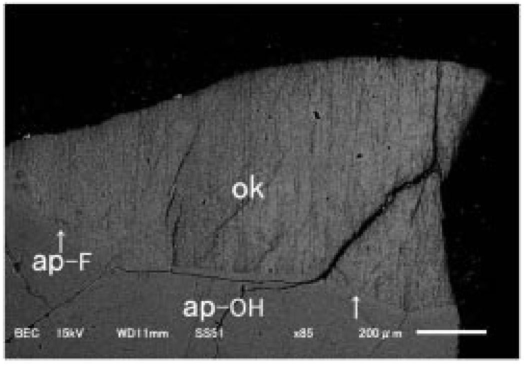

Fig. 10. Back-scattered electron image of okenite (ok), apophyllite-(KF) (ap-F: arrows indicate narrow band contacted with "okenite") and apophyllite-(KOH) (ap-OH).

sideration is interrupted because of few chemical analyses of truscottite. About fluorine content in the present "okenite" we think it is probably significant, because suitable fluorine analyses were resulted for associated apophyllite analyzed at the same time (Fig. 10). As above mentioned the crystal structure of the present "okenite" is probably different with typical okenite. This may be derived from Ca-poor and alkali-rich character in comparison with other okenite. We need more detailed study to establish the "okenite" as a new mineral.

Acknowledgements

We thank Dr. Yuko Uchio, National Museum of Nature and Science, and Mr. Hiroyuki Imai for their help to study and collecting samples in field works. We are also grateful to Mrs. Masako Shigeoka, National Museum of Nature and Science, for her help in EDS analyses.

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