LETTER

Secondary phosphates in montebrasite and amblygonite from Nagatare, Fukuoka Prefecture, Japan

Yohei SHIROSE and Seiichiro UEHARA

Department of Earth and Planetary Sciences, Faculty of Sciences, Kyushu University, Fukuoka 812–8581, Japan

Montebrasite and amblygonite in an Li–Cs–Ta enriched (LCT) pegmatite from Nagatare, Fukuoka Prefecture, Japan, contain various alteration minerals: fluorapatite, crandallite, goyazite, waylandite, wardite, viitaniemiite, morinite, muscovite, lepidolite, and cookeite. They are associated with lacroixite, quartz, and topaz. Among these minerals, wardite, viitaniemiite, morinite, and lacroixite are newly discovered in Japan. The secondary phosphates and fine-grained mica form fine veins along cleavages and composition planes of polysynthetic twins in the montebrasite and amblygonite. Lacroixite has a different texture from other phosphates, which suggests a possibility of exsolution within montebrasite–amblygonite series. Various secondary phosphates show Ca-, Na- and Sr-metasomatism with leaching of Li, and the formation of low-F montebrasite from montebrasite–amblygonite series indicates an F–OH exchange. However, fluorapatite, morinite, and viitaniemiite crystallized in an F-rich environment. Montebrasite–amblygonite series minerals undergo an acidic alteration to muscovite in the last stage, which is the same process that other Li minerals undergo, such as tourmaline and spodumene.

Keywords: Montebrasite–amblygonite series, Viitaniemiite, Morinite, Wardite, Lacroixite

INTRODUCTION

Minerals of the montebrasite–amblygonite series have complete solid solution with $\text{LiAlPO}_4(\text{OH})$ and LiAlPO_4F . Minerals of this series generally occur as primary phosphates from Li–Cs–Ta enriched (LCT) (Černý and Ercit, 2005) pegmatites and topaz-bearing granites. Hydrothermal alteration of the montebrasite–amblygonite series to secondary phosphates during the late stages of pegmatite formation has been recorded at several locations (e.g., London and Burt, 1982; Baldwin et al., 2000; Galliski et al., 2012). In Japan, montebrasite–amblygonite series minerals were first described from an LCT pegmatite in Nagatare, Fukuoka Prefecture. Descriptions were based on optical properties observed by Shibata (1934), and chemical composition and X-ray diffraction data of montebrasite were reported by Ito et al. (1955). The occurrence of montebrasite and alteration to muscovite were also reported at Myokenzan, Ibaraki Prefecture (Sakurai et al., 1977). We have been reinvestigating the minerals of the Nagatare pegmatite including lep-

idolite (Kataoka and Uehara, 2000), tourmaline (Shirose and Uehara, 2013), and other rare minerals, for example, Bi minerals (Uehara and Shirose, 2013). In this study, montebrasite and amblygonite from the Nagatare pegmatite occur with various alteration minerals: muscovite, lepidolite, cookeite, and secondary phosphates such as fluorapatite, crandallite, goyazite, waylandite, wardite $[\text{NaAl}_3(\text{PO}_4)_2(\text{OH})_4 \cdot 2\text{H}_2\text{O}]$, viitaniemiite $[\text{Na}(\text{Ca}, \text{Mn})\text{Al}(\text{PO}_4)\text{F}_2(\text{OH})]$, and morinite $[\text{NaCa}_2\text{Al}_2(\text{PO}_4)_2\text{F}_4(\text{OH}) \cdot 2\text{H}_2\text{O}]$, associated with lacroixite $[\text{NaAl}(\text{PO}_4)\text{F}]$, quartz, and topaz. These phosphates generally occur as hydrothermal products in some pegmatites. This is the first discovery of wardite, viitaniemiite, morinite, and lacroixite in Japan, and this paper describes the occurrence and mineralogical properties of these phosphates.

GEOLOGY AND SAMPLE DESCRIPTIONS

The Nagatare pegmatite is an LCT pegmatite, located at Nagatare, in the western area of Fukuoka City, Fukuoka Prefecture. The pegmatite is derived from the Sawara granite (80–100 Ma) that intruded into the Itoshima granodiorite (70–115 Ma) (Karakida et al., 1994). There are outcrops of barren pegmatite and beryl-containing peg-

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Y. Shirose, shirose@kyudai.jp Corresponding author

S. Uehara, uehara@geo.kyushu-u.ac.jp

Table 1. Sample numbers with the associated minerals and the properties of montebrasite and amblygonite

Sample no.	Size (cm)	Form	Color	Associated minerals*	F contents** (apfu)	Phosphates							
						Ap	Cdl	Gyz	Vtm	Mor	Wd	Lcx	Bi
XN162	6	Block	LS-CL-MW	Qtz, Lpd, Ab	0.59	○	○	○				○	
XN165	2	Block	LS-MW	Ab, Qtz	0.56	○	○	○				○	○
XN160	4	Block	CL-MW	(Qtz, Lpd, Ab)	0.54	○	○	○	○	○	○	○	○
XN163	4	Block	CL-LS-MW	Qtz	0.54	○	○	○	○	○		○	
XN168	0.5	Dendritic	LS-CL-MW	Kfs, Qtz, Lpd, Ab, (Tur)	0.50	○	○	○			○	○	○
Ng104	0.5	Dendritic	CL-MW	Kfs, Qtz, Lpd, (Ab, Tur)	0.48	○	○	○			○	○	
XN167	0.3	Block	CL-MW	Qtz, Lpd, Ab, Kfs, Tur	0.48	○	○	○				○	
XN169	1	Block	LS-MW	Ab, (Qtz, Lpd)	0.47	○	○					○	
Ng107	4	Block	MW	(Qtz, Lpd)	0.44	○	○		○		○	○	
XN164	2	Block	LS-MW	Ab, Ptl, (Lpd)	0.41	○	○					○	
XN166	2	Block	MW	Qtz, Lpd-Ms, (Ab)	0.40	○	○					○	

* Slightly associated minerals are noted in brackets.

** F contents were estimated from bulk powder XRD patterns for all specimens using Kallio's (1978) method.

LS, light-salmon color; CL, colorless; MW, milky-white color. Qtz, quartz; Lpd, lepidolite; Ab, albite; Kfs, K-feldspar; Tur, Li tourmaline; Ptl, petalite; Ms, muscovite; Ap, fluorapatite; Cdl, crandallite; Gyz, goyazite; Vtm, viitaniemiite; Mor, Morinite; Wd, wardite; Lcx, lacroixite; Bi, Bi phosphates.

Figure 1. Appearance and optical photomicrograph of montebrasite and amblygonite. (a) and (b) Photographs of subhedral amblygonite from the Nagatare pegmatite (samples XN162 and XN163). (c) Photograph of dendritic montebrasite in K-feldspar from the Nagatare pegmatite (sample Ng104). (d) Cross section of amblygonite (sample XN162). Arrows represent the white veins parallel to the cleavages. (e) and (f) Plane- and cross-polarized optical photomicrographs showing the cloudy parts and the polysynthetic twins of amblygonite (sample XN160). Amb, amblygonite; Mtb, montebrasite; Lpd, lepidolite; Ab, albite; Qtz, quartz; Kfs, K-feldspar.

matite found from the coast to Mt. Nagatare. The Li-enriched pegmatite of Mt. Nagatare was mined for Li ore in the middle of the 20th century. The ore body contained rare element minerals such as lepidolite, Li tourmaline, petalite, montebrasite, amblygonite, and pollucite. Eleven specimens used in this study were collected from this old ore deposit (Table 1). The sample numbers are listed with the results of the analyses in Table 1.

The montebrasite and amblygonite from the Nagatare pegmatite occur as large subhedral to euhedral crystals with lepidolite, Li tourmaline and petalite in quartz, and albite (Figs. 1a and 1b). Small dendritic crystals of montebrasite are contained within K-feldspar (Fig. 1c).

The crystals are colorless, milky-white, or light-salmon, and have a vitreous luster. There are white veins parallel to cleavage planes in the cross sections (Fig. 1d). Under the polarizing microscope, montebrasite and amblygonite were mostly cloudy caused by very fine inclusions, and polysynthetic twins were observed (Figs. 1e and 1f). Manganotantalite, microlite, and minor bismuth were also observed as inclusions within montebrasite and amblygonite.

ANALYTICAL METHODS

Chemical analyses were performed on a JEOL

Figure 2. BSE images of the secondary phosphates in the montebrasite and amblygonite. (a) and (b) Crandallite and apatite forming veinlets along cleavages and composition planes of polysynthetic twins in montebrasite (sample Ng107). (c) Chemical zoning texture of prismatic crandallite-goyazite series minerals (sample XN160). (d) Viitaniemiite with high fluorine contents silicates (sample XN160). (e) Morinite with quartz (sample XN163). (f) Wardite with veinlets of other phosphates (sample XN160). Mtb, montebrasite; Amb, amblygonite; Cdl, crandallite; Ap, fluorapatite; Gyz, goyazite; Tpz, topaz; Vtm, viitaniemiite; Qtz, quartz; Lpd, lepidolite; Wdt, wardite; Mor, morinite.

JXA8530F electron microprobe (EPMA) equipped with a wavelength dispersive X-ray spectrometer. Quantitative analyses were performed at an accelerating voltage of 15 kV, beam current of 2 nA, and probe diameter of 5–15 μm . The standard materials were fluorapatite (for $\text{FK}\alpha$, $\text{PK}\alpha$, and $\text{CaK}\alpha$), albite (for $\text{NaK}\alpha$), forsterite (for $\text{MgK}\alpha$), almandine (for $\text{AlK}\alpha$ and $\text{FeK}\alpha$), celestite (for $\text{SK}\alpha$ and $\text{SrL}\alpha$), and rhodonite (for $\text{MnK}\alpha$). Analyses of montebrasite and amblygonite were performed with the other appropriate standard set: fluorophlogopite (for $\text{FK}\alpha$), spodumene (for $\text{AlK}\alpha$), KTiOPO_4 (for $\text{PK}\alpha$), albite (for $\text{NaK}\alpha$), diopside ($\text{CaK}\alpha$), and almandine (for $\text{FeK}\alpha$). This standard set was chosen to increase accuracy by using standards with similar concentrations to the specimens. The ZAF method was used for data correction. Samples of the crandallite-goyazite series and wardite suffered extensive damage from the electron beam. X-ray diffraction (XRD) data was collected on crystal fragments using a Rigaku RINT RAPIDII curved imaging plate microdiffractometer that used monochromatized $\text{CuK}\alpha$ radiation generated at 40 kV and 30 mA. The fragments were randomized using a Gandolfi-like motion about two axes (oscillation on ω and rotation on ϕ). The XRD data of crandallite-goyazite series, viitaniemiite, morinite, and wardite were referenced using PDF cards: #01-086-2315 (Kato, 1971), #00-035-0598 (Ramik et al., 1983), #01-076-1881 (Hawthorne, 1979), and #01-070-2154 (Fanfani et al., 1970), respectively.

DESCRIPTION OF MINERALS

Macroscopic white veins correspond to the veins of fine-grained secondary minerals (Fig. 1d). The thin veins are under 400 μm in width, found along cleavages and composition planes of polysynthetic twins in montebrasite and amblygonite, and composed of fine-grained mica and secondary phosphates (Fig. 2). Phosphates were sort-

ed into Ca phosphates, Na-Ca phosphates, and Na phosphates. The chemical compositions and the XRD data are in Table 2 and Appendix tables 1–4, respectively. Fluorine contents for montebrasite-amblygonite series minerals were estimated from bulk powder XRD patterns for all specimens using Kallio's method (1978). The obtained values of $\text{F}/(\text{F} + \text{OH})$ in the bulk compositions are 0.40–0.59 (Table 1). However, EPMA analyses show they are partially replaced by low-F montebrasite (Fig. 3 and Table 3). The detailed mineralogical studies of montebrasite, amblygonite, and lacroixite will be reported in another paper (Shirose and Uehara, in prep.).

Ca phosphates

Fluorapatite and crandallite are found in all specimens (Table 1). Fluorapatite forms veinlets composed of radial aggregates of acicular crystals (Figs. 2a and 2b). Grained single crystals of fluorapatite are also included in montebrasite and amblygonite. While some grained fluorapatites contain Mn and Sr, acicular fluorapatite has relatively poor Mn content and no Sr content. Regarding F content, the fluorapatite grains are mostly fluorapatite end-member, and some hydroxylapatites exist with muscovite. Crandallite occurs in acicular to tabular aggregates as veinlets (Figs. 2a and 2b) and contains Sr in some specimens (Table 2). Goyazite, the Sr analogue of crandallite, also occurs with crandallite (Fig. 2c and Table 2) and exhibits compositional zoning textures. The unit cell parameters of sample XN162 are close to that of goyazite, reflecting the composition (Appendix table 1). The Nagatare goyazite is the first Sr mineral found in the pegmatite.

Na-Ca phosphates

Viitaniemiite and morinite are high-F species compared

Table 2. Chemical composition of the phosphates in the montebrasite and amblygonite from the Nagatare pegmatite

Species	Crandallite		Goyazite	Fluorapatite	Viitaniemiite		Morinite	Wardite	Lacroixite
Sample no.	Ng107	XN166	XN163	XN164	XN163	XN160	XN163	XN160	XN163
n*	3	1	2	3	3	5	3	4	2
SO ₃	0.7 (4)	0.1	-	0.0 (0)	-	-	-	-	-
P ₂ O ₅	33.2 (1)	32.3	29.2 (7)	43.1 (7)	31.4 (3)	28.7 (9)	30.5 (2)	36.2 (8)	45.5 (2)
Al ₂ O ₃	32.6 (3)	30.8	30.0 (1)	0.3 (2)	20.2 (5)	20.4 (3)	19.9 (4)	34.6 (3)	29.4 (0)
MgO	-	-	-	-	2.2 (13)	0.2 (1)	-	-	-
CaO	13.0 (3)	8.1	0.2 (1)	56.0 (6)	20.9 (24)	21.8 (6)	23.2 (4)	0.2 (1)	0.4 (3)
MnO	-	-	-	0.9 (6)	0.4 (2)	1.2 (4)	-	-	-
SrO	0.0 (1)	7.0	19.2 (3)	0.0 (0)	-	-	-	-	-
Na ₂ O	0.0 (0)	0.0	-	0.0 (0)	12.7 (2)	11.8 (3)	6.2 (1)	7.1 (1)	16.8 (3)
F	1.9 (3)	1.0	1.6 (1)	3.9 (4)	13.0 (8)	13.6 (5)	12.8 (3)	1.6 (7)	10.4 (3)
H ₂ O**	13.4	13.1	11.9	0.0	5.2	4.4	10.7	16.6	0.6
O=F	0.8	0.4	0.7	1.6	5.5	5.7	5.4	0.7	4.4
Total	94.2	92.0	91.4	102.7	99.9	96.5	98.1	95.7	98.7
	O = 10.5		O = 10.5	O = 12.5	O = 5.5		O = 10.5	O = 10.0	O = 4.5
S apfu	0.04 (2)	0.01	-	0.00	-	-	-	-	-
P	2.06 (1)	2.12	2.06 (2)	2.99 (2)	1.05 (1)	1.00 (1)	2.07 (1)	2.12 (2)	1.05 (0)
Al	2.81 (3)	2.81	2.94 (4)	0.03 (2)	0.94 (2)	1.00 (2)	1.88 (3)	2.81 (3)	0.95 (1)
Mg	-	-	-	-	0.08 (9)	0.01 (9)	-	-	-
Ca	1.02 (2)	0.67	0.02 (1)	4.92 (9)	0.89 (11)	0.97 (11)	2.00 (3)	0.01 (1)	0.01 (1)
Mn	-	-	-	0.06 (4)	0.01 (1)	0.04 (1)	-	-	-
Sr	0.00 (0)	0.32	0.93 (0)	0.00 (0)	-	-	-	-	-
Na	0.00 (0)	0.00	-	0.00 (0)	0.97 (2)	0.95 (2)	0.97 (2)	0.96 (2)	0.89 (1)
F	0.44 (8)	0.25	0.43 (4)	1.01 (10)	1.63 (9)	1.78 (9)	3.24 (7)	0.34 (15)	0.90 (2)
OH	4.56 (8)	4.75	4.57 (4)	0.00	1.37	1.22	1.76 (7)	3.66 (15)	0.10 (2)
Cation sum	5.93	5.91	5.94	8.00	3.94	3.97	6.93	5.90	2.89

* Number of analyzed spots.

** H₂O contents were calculated by stoichiometry. **Figure 3.** Plane- and cross-polarized optical photomicrograph and X-ray images of elements distribution map (NaK α , FK α , PK α , and AlK α), indicating the replacement by low-F montebrasite (sample XN165). The part with blue interference-color is secondary montebrasite similar to London and Burt (1982), and it has low F content. Ab, albite; Amb, amblygonite; Mtb², secondary low-F montebrasite; Lcx, lacroixite.

to other phosphates, and they are present in a few specimens as veinlets composed of micro-tabular crystals (Figs. 2d and 2e). In this locality, viitaniemiite is usually associated with lepidolite and topaz, and contains Mn and Mg as minor elements (Table 2). The chemical compositions and unit cell parameters are more similar to Mn-free

viitaniemiite (Ramik et al., 1983; Breiter et al., 2009) than Mn-bearing (up to 0.4 apfu) viitaniemiite (Lathi, 1981; Petrik et al., 2011) (Appendix table 2). The veinlets of morinite include fluorapatite and zoned crystals of the crandallite-goyazite series, extending 2 cm in length and 20 μ m in width. Morinite has no Mn or Mg and has slightly lower F than the ideal formula as with the viitaniemiite (Table 2). The unit cell parameters are in good agreement with those of morinite (Hawthorne, 1979) (Appendix table 3).

Na phosphates

Wardite occurs in veinlets like the other phosphates, but these are cut by or shifted into Ca phosphate and Na-Ca phosphate (Fig. 2f). Wardite from Nagatare has slightly larger unit cell parameters than that from Fairfield (Fanfani et al., 1970; Appendix table 4), and this difference is caused by slightly-contained F (Table 2). Lacroixite is found in all specimens as scattered lamella embedded in montebrasite and amblygonite, unlike other secondary phosphates (Figs. 3 and 4). This texture is similar to 'natromontebrasite', a discredited species that was deter-

Table 3. Representative chemical composition of the amblygonite, montebrasite, and low-F montebrasite from the Nagatare pegmatite

Species	Amblygonite	Montebrasite	Low-F montebrasite
Sample no.	XN160	XN166	XN165
P ₂ O ₅	47.72	48.33	48.16
Al ₂ O ₃	34.53	34.61	35.04
CaO	0.00	0.02	0.03
FeO	0.01	0.00	0.00
Li ₂ O*	10.07	10.16	10.19
Na ₂ O	0.00	0.00	0.01
F	7.12	3.84	1.36
H ₂ O*	2.70	4.31	5.50
O=F	3.00	1.62	0.57
Total	99.15	99.64	99.71
		O = 4.5	
P	1.00	1.00	0.99
Al	1.00	1.00	1.01
Ca	0.00	0.00	0.00
Fe	0.00	0.00	0.00
Li	1.00	1.00	1.00
Na	0.00	0.00	0.00
F	0.56	0.30	0.10
OH	0.44	0.70	0.90
Cation sum	3.00	3.00	3.00

* Li₂O and H₂O contents were calculated by stoichiometry.

Figure 4. Scattered lamella of lacroixite embedded in montebrasite (sample Ng107). Mtb, montebrasite; Lcx, lacroixite.

mined to be a mixture of an OH-rich amblygonite and lacroixite (Fransolet, 1989; Fransolet et al., 2007). The montebrasite-amblygonite series is topologically identical to the lacroixite structure, and Na could not substitute Li at low temperatures in the montebrasite-amblygonite structure (Groat et al., 1990) (Table 3). Thus, it is possible that lacroixite is exsolved as a Na-bearing phase as the temperature decreased and is subsequently distributed within montebrasite and amblygonite.

DISCUSSION

Montebrasite and amblygonite from the Nagatare pegmatite underwent staged metasomatic alteration. The formation of low-F montebrasite from montebrasite and amblygonite indicates an F-OH exchange, and various secondary phosphates show Ca-, Na- and Sr-metasomatism,

Figure 5. General alteration sequence of montebrasite-amblygonite series without Fe and Mn phosphates. Asterisk represents the minerals that occurred in the Nagatare pegmatite.

with leaching of Li. However, the existence of fluorapatite, morinite, and viitaniemiite indicates that they formed in an F-rich environment. The low Mn content of viitaniemiite and the lack of Fe and Mn phosphates as replacements of montebrasite and amblygonite indicate that these secondary phosphates crystallized in an Fe- and Mn-free environment differing from the other stages [e.g., triplite occurring stage (Uehara et al., 2010)] in the Nagatare pegmatite.

A sequence of metasomatic alterations of the montebrasite-amblygonite series minerals from the pegmatites of the White Picacho District was established by London and Burt (1982) in the following order: montebrasite → low-F montebrasite → hydroxylapatite + crandallite → hydroxylapatite + muscovite + brazilianite + augelite + Fe and Mn phosphates → muscovite → carbonate-apatite. Similar sequences are reported from other pegmatites (e.g., Baldwin et al., 2000; Galliski et al., 2012) where goyazite and gorceixite are included, and Fe and Mn phosphates are absent, unlike in the case of the White Picacho District. The Nagatare pegmatite displays characteristic wardite, not brazilianite, morinite, and viitaniemiite formation. Integration of the previous sequences and that of the Nagatare pegmatite results in a general alteration sequence of montebrasite-amblygonite series minerals without Fe and Mn phosphates (Fig. 5). Lacroixite has a different texture than other phosphates, and this indicates a possibility of exsolution within the montebrasite-amblygonite series. Crandallite generally occurs as a secondary phosphate in the montebrasite-amblygonite series, and the same group of minerals also occurs depending on the chemical environments, which behaves as a sensitive chemical indicator corresponding to the chemical compositions of the solid solutions. The montebrasite-amblygonite series minerals undergo an acidic alteration to muscovite in the last stage, which is the same process that other Li minerals undergo,

such as tourmaline (Ahn and Buseck, 1998) and spodumene (London and Burt, 1982).

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SUPPLEMENTARY MATERIALS

Appendixes tables 1–4 are available online from <http://japanlinkcenter.org/DN/JST.JSTAGE/jmps/131022d>.

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