Cl and F contents of apatite in the Matsumae plutonic rocks, southwestern Hokkaido, Japan — A useful indicator of vapor saturation —

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Chlorine and fluorine contents of apatite in the Matsumae plutonic rocks were analyzed with electron microprobe. The Matsumae plutonic rocks are divided into Am and Pl series in terms of crystallization sequence and mineralogy. The petrological evidence suggests that the Am series rocks have crystallized under the conditions richer in water than the Pl series. Chlorine content of apatite in the Am series is generally low, whereas that in the Pl series is rather high in less differentiated rocks and abruptly decreases at about D.I.=45. On the other hand, fluorine content of apatite in both series slightly increases as differentiation proceeds.

Such behavior of Cl and F in apatite can be explained in terms of the degassing process; when a magma is saturated with vapor and exsolves free vapor phase, Cl is more strongly partitioned into the vapor relative to F and removed from the magma. Therefore, judging from low Cl content of apatite, the Am series magma may have been saturated with vapor in earlier stage than the Pl series one. This is consistent with the previous suggestion that the Am series magma have crystallized under the conditions richer in water than the Pl series. Such abrupt decrease in Cl without decrease in F may be a useful indicator of vapor exsolution or vapor saturation of magma. Recognition of vapor saturation gives valuable information on water content of magma.

Introduction

The Matsumae plutonic rocks are divided into Am and Pl series: the Am series is characterized by crystallization of amphibole prior to plagioclase, whereas the Pl series by reverse crystallization sequence. The Am series magma may have contained more water than the Pl series magma (Tsuchiya, 1985a). However, the variation in oxidation state between the two series magmas does not strictly correlate with such difference in supposed water content (Tsuchiya, 1985b). This fact is not consistent with the explanation that oxidation of magma is brought about by separation of an aqueous phase from the magma (e.g. Czamanske *et al.*, 1981). The investigation of Cl and F

in magma has great value in elucidating the role of vapor phase.

Cl and F are generally contained in OH-bearing minerals such as mica, amphibole, and apatite. Among them, apatite is a common accessory mineral which is contained in every rock type of the Matsumae plutonic rocks. In addition, Cl in apatite may be less affected by postmagmatic alteration than that in mica (e. g., Roegge *et al.*, 1974; Nedachi, 1980). Therefore, apatite is the best mineral to understand the behavior of halogen concentration in magma. In this paper, Cl and F contents of apatite are described and the role of vapor phase in the Matsumae plutonic magma is discussed.

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Petrological outline

The Matsumae plutonic rocks consist of various rock types from ultramafic cumulates to adamellite. The clinopyroxenite, hornblendite I, II, granodiorite, and aplite I belong to the Am series, whereas the monzogabbro, quartz monzodiorite, adamellite, and aplite II to the Pl series. Differentiation trend of the Am series is explained by the amphibole-dominant fractionation, whereas that of the Pl series by the fractionation of plagioclase plus mafic minerals. Both series rocks are characteristically high in alkali, especially in K_2O content, though the Am series rocks are slightly less alkalic than the Pl series. Petrological and mineralogical evidence suggests that the Am series magma contained more water than the Pl series (Tsuchiya, 1985a). Variation in oxidation state of the Matsumae plutonic magma is not strictly correlate with this suggestion of water content. Namely, the Pl series rocks are characterized by high oxidation state of magma, whereas the Am series rocks show various oxidation state. Detailed study of the Matsumae plutonic rocks is described by Tsuchiya (1982, 1985a, b).

Occurrence of apatite

Apatite is contained in every rock type, and its modal composition varies to a more extent in the Am series than in the Pl series. In the clinopyroxenite, only a trace amounts of apatite is present, and it occurs as anhedral to subhedral intercumulus crystals up to 2 mm. The apatite in the hornblendite I shows conspicuous modal variation (0 to 5 vol%), and generally occurs as euhedral cumulus crystals (10 to 100 μm) included in amphibole, biotite, plagioclase, K-feldspar, and quartz. The apatite in the granodiorite is euhedral (5 to 100 μm), and its modal abundance ranges from 0.1 to 3 vol%.

Apatite content of the Pl series rocks is

generally less than 3 vol%, and shows no considerable variation among the rock types. The apatite usually occurs as euhedral crystals up to 200 μm . The quartz monzodiorite and fine-grained monzogabbro occasionally contain small needle-like crystals of apatite (less than $1 \times 20 \mu m$). These apatites in the Pl series are usually included in amphibole, biotite, plagioclase, K-feldspar, and quartz. In some specimens of the monzogabbro and quartz monzodiorite, apatite is rarely included in clinopyroxene.

These apatites are almost colorless under the microscope, but some apatites in the hornblendite I and granodiorite are pale brown.

Chemistry of apatite

Cl and F in apatite were analyzed using the

Table 1. Representative analyses of apatite

	HOR I 52706C	QMD 80602	QMD 52901
P ₂ O ₅	42.28	41.18	42.70
SiO ₂	.16	.27	.47
TiO ₂	.03	tr	.03
Al ₂ O ₃	.01	.01	.03
FeO*	.25	.31	.40
MnO	.08	.08	.13
MgO	.18	.03	.02
CaO	54.44	55.15	54.63
Na ₂ O	.09	.09	.17
F	1.35	1.88	2.34
Cl	.30	.91	.34
H ₂ O	1.07	.65	.61
Sum	100.24	100.56	101.87
-O=F, Cl	.64	1.00	1.06
Total	99.60	99.56	100.81
Structural formulae on the basis of 26 (O, OH, F, Cl)			
P	6.013	5.918	6.002
Si	.027	.046	.078
Ti	.004	-	.004
Al	.002	.002	.006
Fe	.035	.044	.056
Mn	.011	.012	.018
Mg	.045	.008	.005
Ca	9.798	10.031	9.718
Na	.029	.030	.055
F	.717	1.009	1.229
Cl	.085	.262	.096
OH	1.198	.729	.675
Total	17.964	18.091	17.942

FeO*: total Fe as FeO, tr: not detected.
HOR I: hornblendite I, QMD: quartz monzodiorite.

Table 2. Analyses of F and Cl in apatite

	rock type	sample No.	Type	F (wt%)		Cl (wt%)		n
				range	average	range	average	
Am series	CPX	52306	II	.53 - 1.22	.79 (.27)	.23 - 1.11	.91 (.31)	7
	HOR I	503R	II	1.90 - 2.52	2.11 (.36)	.08 - .34	.20 (.13)	3
		52706	I-II	1.50 - 2.84	1.92 (.37)	.00 - .34	.15 (.12)	18
		52706C	I	1.35 - 2.84	1.92 (.45)	.14 - .34	.25 (.05)	15
		52303	II	1.94 - 2.52	2.26 (.22)	.15 - .22	.19 (.02)	8
		91611	I	1.69 - 2.06	1.88 (.11)	.10 - .22	.17 (.03)	10
	HOR II	91708	II	.82 - 2.50	1.47 (.56)	.09 - .75	.43 (.17)	22
	GD	62810	II	1.31 - 2.25	1.70 (.40)	.11 - .49	.23 (.13)	8
		71904	II	1.77 - 2.59	2.10 (.24)	.31 - .58	.43 (.09)	9
		91802	III	1.73 - 2.35	1.94 (.21)	.00 - .09	.02 (.03)	10
		82108	I	1.94 - 3.08	2.51 (.36)	.02 - .19	.07 (.06)	9
		52617	I	1.93 - 2.48	2.32 (.22)	.00 - .04	.02 (.02)	6
Pl series	MG	52210	I	1.16 - 1.61	1.40 (.12)	.79 - 1.02	.91 (.08)	10
		82104	II	2.25 - 2.32	2.29 (.04)	.73 - .91	.84 (.10)	3
	QMD	80602	I	1.63 - 2.40	2.00 (.24)	.80 - 1.11	.98 (.12)	12
		80716	I	1.38 - 1.88	1.64 (.20)	1.11 - 1.28	1.19 (.07)	5
		80713	I	1.84 - 2.84	2.52 (.27)	1.03 - 1.23	1.13 (.08)	11
		80903	I	2.07 - 2.50	2.27 (.15)	.35 - .51	.42 (.04)	11
		52901	I	2.25 - 2.84	2.52 (.27)	.24 - .34	.28 (.05)	5
		52907	I	1.31 - 2.01	1.72 (.21)	.22 - .42	.30 (.07)	7
		80907	I	2.02 - 2.79	2.48 (.34)	.08 - .21	.12 (.04)	8
	AD	91505	I	2.08 - 2.79	2.42 (.25)	.01 - .06	.04 (.02)	5
		52109	I	1.89 - 3.04	2.58 (.45)	.01 - .06	.03 (.02)	7

CPX, clinopyroxenite; HOR I, hornblende I; HOR II, hornblende II; GD, granodiorite; MG, monzogabbro; QMD, quartz monzodiorite; AD, adamellite. These host rocks are classified into Type I to III in terms of the characteristics of Fe-Ti oxide minerals (Tsuchiya, 1985b).

Figures in parenthesis are standard deviation. n, number of analyses.

electron microprobe JEOL JXA-50A in Department of Metallurgical Engineering, Hokkaido University. Excitation voltage of 15 kv, sample current of 0.02 to 0.03 μ A on apatite, integration time of 70 seconds, and a beam diameter of 10 μ m were used. Analyzing crystal was RAP for F and PET for Cl. Natural fluorite and fluorapatite were used as standards for F, and a pure reagent of NaCl was used for Cl. A correction was made by the method of Bence and Albee (1968) and used an alpha factor of Albee and Ray (1970) assuming stoichiometric values of CaO and P₂O₅. In some cases, P, Si, Ti, Al, Fe, Mn, Mg, Ca, and Na were also analyzed. Analytical error is estimated to be

15 % for F and 4 % for Cl in terms of standard deviation of each value of raw counts.

Table 1 shows representative analyses of apatite in three specimens. H₂O content is calculated assuming F+Cl+OH=2.0 in the structural formulae on the basis of O=25. These apatites are almost stoichiometric, and probably contain only trace amounts of other elements (e.g. C, S, and REE). SiO₂ and FeO are detected up to 0.5 and 0.4 wt%, respectively. Several authors reported similar values for such components (e.g. Nash, 1976). Except for halogen content, there is no significant difference in chemical composition of apatite from specimen to specimen. There is no corre-

Fig. 1. Molecular proportions of F, OH, and Cl for apatite. Abbreviations of rock types are shown in Table 2. Ichinohe, apatites from the Ichinohe plutonic complex; OM, orthopyroxene-olivine-biotite-clinopyroxene monzonite; HM, biotite-hornblende monzonite.

lation between the color of apatite and its chemistry.

Table 2 lists Cl and F contents of apatite in each rock type, and Fig. 1 shows molecular proportion of F, OH, and Cl. The apatite in the Am series rocks is generally poor in Cl, except for that in the clinopyroxenite which is relatively rich in Cl and poor in F. In the hornblendite II and granodiorite, the apatite included in the core of amphibole is richer in Cl than that included in the rim of amphibole and that in the interstices. Such occurrence indicates that Cl content is slightly richer in the early-crystallized apatite than in the late-crystallized one. F content of the apatite in the Am series rocks shows a wide variation even within a single specimen.

The apatite in the Pl series rocks shows a

wide range of Cl content; the apatite in the monzogabbro and some of the quartz monzodiorite is richer in Cl than that in the rest of the Pl series rocks. F content of the apatite in the Pl series rocks shows a wide variation within a single specimen, but there is no significant difference among the rock types.

Apatite in the Ichinohe plutonic rocks in the Kitakami belt, which have a similar petrochemical character to the Matsumae plutonic rocks, was analyzed and is also plotted in Fig. 1 for comparison. The apatite in the orthopyroxene-olivine-biotite-clinopyroxene monzonite (kentallenite) is rich in Cl (1.12 to 1.77 wt%), whereas that in the biotite-hornblende monzonite is poor in Cl (0.21 to 0.43 wt%). The apatite in the latter rock shows a rather wide range of F content.

Fig. 2. F and Cl contents of apatite vs. D.I. of the host rocks except for the cumulates. Vertical bar represents 2σ . Note the abrupt decrease in Cl content of apatite in the Pl series at about D.I. = 45.

Fig. 2 shows Cl and F contents of apatite versus D.I. of the host rocks except for the cumulates. F content of the apatite shows no significant difference between the Am and Pl series, and slightly increases with increasing D.I.. On the other hand, behavior of Cl in apatite shows marked difference between the two series. In the Am series, Cl content of apatite is generally low, though it is slightly higher in less-differentiated rocks. In the Pl series, Cl content of apatite is rather high and slightly increases with increasing D.I. within a range of low D.I., and abruptly decreases at about D.I. = 45.

In Table 2, the host rocks are divided into

three types in terms of the characteristics of Fe-Ti oxide minerals: Type I is characterized by the presence of magnetite and hemo-ilmenite, Type II by the presence of magnetite and absence of hemo-ilmenite, and Type III by the absence of magnetite. Among them, Type I indicates the crystallization at highest oxidation state, whereas Type III at lowest (Tsuchiya, 1985b). As shown in Table 2, no correlation is recognized between the behavior of Cl content of apatite and the oxidation state of the host magma.

Discussion

1. Different behavior of Cl in apatite between

the two series

The previous workers showed that the F content of igneous rocks generally increases with differentiation, whereas the Cl content decreases or shows no correlation with differentiation (e.g. Banks, 1976; Fuge, 1977). The behavior of Cl and F in apatite in the Matsumae plutonic rocks shows a similar tendency, but there is a distinct difference in the behavior of Cl between the Am and Pl series. The Pl series rocks are characterized by the abrupt decrease in Cl in apatite at about D.I.=45 as described above.

If the distribution coefficient of Cl and F between apatite and magma has not greatly changed during differentiation, the concentration of these elements in apatite must correlate with that in magma. Therefore, decreasing trend of Cl in apatite indicates decrease in Cl in magma, which seems to result from fractionation of Cl-bearing minerals or degassing (e.g. Anderson, 1974; Muramatsu and Wedepohl, 1979). Principal Cl-bearing minerals in the Matsumae plutonic rocks are apatite, amphibole, and biotite, and Cl content of the rest minerals is negligible. According to Anderson (1974), Cl content of such minerals is not so high and Cl cannot be effectively extracted from magma by fractional crystallization. In the case of the Matsumae plutonic rocks, maximum Cl contents of amphibole and biotite are 0.3 and 0.4 wt%, respectively (Tsuchiya, unpublished data). However, there is no evidence of appearance or abrupt increase in modal abundance of such Cl-bearing minerals in the Pl series at about D.I.=45. In addition, decrease in F content, which is expected to result from fractionation of such Cl-bearing minerals, is not observed in apatite in the Pl series rocks. Therefore, the abrupt decrease in Cl in apatite cannot be explained by the fractionation of Cl-bearing minerals, but by the degassing.

If the magma becomes saturated with vapor, vapor separates from the residual magma by a process commonly called second or retrograde boiling (e.g. Burnham and Ohmoto, 1980). Exsolved vapor phase probably acts as a carrier gas of Cl because of strong affinity of Cl for the aqueous vapor phase (Kilinc and Burnham, 1972). On the other hand, since most of F would tend to remain in magma under such conditions (Munoz and Eugster, 1969), F content may not be strongly affected by vapor exsolution. Therefore, vapor exsolution results in abrupt decrease in Cl content of magma without decrease in F. Namely, the Pl series magma may have led to vapor saturation and exsolved free vapor phase at about D.I.=45. Consequently, the abrupt decrease in Cl without decrease in F can be a useful indicator of vapor exsolution or vapor saturation.

Anderson (1974) stated that water-rich magma may lose most of their Cl by degassing in relatively early stage of crystallization. In the case of the Am series rocks, therefore, low Cl content of apatite suggests that the Am series magma was saturated with vapor in early stage of crystallization. This consists with the idea that the Am series magma contained more water than the Pl series one (Tsuchiya, 1985a).

Abrupt decrease in Cl without decrease in F as described above is also recognized in some plutonic rocks in the Kitakami belt. In the Ichinohe plutonic rocks, for example, the apatite contained in less differentiated rock (orthopyroxene-olivine-biotite-clinopyroxene monzonite) is rich in Cl, suggesting that the magma was undersaturated with vapor. On the other hand, more differentiated rock (biotite-hornblende monzonite) seems to be saturated with vapor because of low Cl and rather high F content of apatite. In addition, Nedachi and Ueno (1981) described a similar

behavior of Cl and F in apatite in the Orikabe granitic rocks. Several authors reported a similar variation of Cl and F in some granitic rocks (e.g. Tsusue *et al.*, 1981, 1984). These data indicate that vapor saturation have occurred at a certain stage of the crystallization of their magma.

Kilinc and Burnham (1972) showed that partition of Cl in an aqueous phase decreases with increasing pressure higher than about 7 kb. Therefore, degassing process discussed above easily occurs at a relatively low pressure range. The depth of intrusion of the Matsumae plutonic rocks can be estimated in terms of the mineralogy of hornfels in the contact aureole. The pelitic hornfels generally composed of plagioclase, quartz, K-feldspar, muscovite, biotite, andalusite, and cordierite with or without minor amounts of sillimanite. The cordierite coexisting with biotite, K-feldspar, andalusite, and muscovite is relatively rich in Fe ($\text{Fe}/\text{Fe} + \text{Mg} = 0.51 - 0.54$; Tsuchiya, unpublished data). This indicates that the Matsumae plutonic rocks have crystallized at a rather low pressure less than 4 kb (Holdaway and Lee, 1977). The degassing process may have been easily occurred at such low pressure conditions.

The estimated oxidation state of the Matsumae plutonic rocks has no correlation with the variation in Cl content of apatite as described above. Moreover, some of the magnetite-series rocks show high Cl content which may indicate vapor undersaturation (e.g., Tsusue *et al.*, 1981; Nedachi and Ueno, 1981). These facts suggest that vapor exsolution does not play a significant role on the oxidation of magma. Therefore, the oxidation of magma may not be brought about by vapor exsolution but probably by diffusive loss of H_2 through the country rocks.

2. Estimation of water content of the Matsumae plutonic magma

In vapor-saturated magma, water content at a given pressure and temperature can be estimated by several methods, assuming that the vapor phase is pure water. The mixing model of albite-water of Burnham (1974, 1979) is adopted here. The Matsumae plutonic rocks may have crystallized at less than 4 kb as described above, and the pressure of crystallization is assumed to be 3 kb. Crystallization temperature of 800°C is arbitrarily chosen. Since the solubility of water in albite melt depends not largely on temperature at a relatively low pressure range (Burnham and Davis, 1974; Burnham and Ohmoto, 1980), it is not necessary to consider the variation in crystallization temperature. On the other hand, confining pressure gives considerable effect on the estimation of water content: if the pressure decreases to 2 kb, calculated water content is lowered about 1.5 wt%. The results of calculation are shown in Fig. 3. There is no significant difference between the two series rocks in water content of the water-saturated magma. Water content obtained here would correspond to a maximum value, because the vapor phase is generally not pure water.

Water content of water-undersaturated magma cannot be estimated by above method. The specimen No. 80602 has the least D.I. among the quartz monzodiorite, and its magma seems to be undersaturated with water judging from the high Cl content of apatite. Magmatic water content generally increases with fractionation of anhydrous minerals. Such phenomenon may occur in fractionation of lesser amounts of hydrous minerals together with anhydrous minerals. Therefore, if the magma is completely closed system concerning with water, the water content of water-undersaturated magma can be estimated by the assemblage and proportion of fractionated

Fig. 3. Estimated water content of the Matsumae plutonic magma.

minerals to yield water saturation.

The quartz monzodiorite magma was originally water-undersaturated (No. 80602), and the water content probably increased with crystallization, which resulted in water-saturation at about D.I.=45 (No. 80903). Therefore, equation of fractionation is assumed as follows; primary magma (No. 80602) = fractionated minerals + residual magma (No. 80903). This equation can be solved by the same method as Tsuchiya (1985a). The result of calculation is; primary magma = $0.0377 \text{ cpx} + 0.2285 \text{ amph} + 0.077 \text{ bio} + 0.0012 \text{ ore} + 0.1518 \text{ pl} + 0.5042 \text{ residual magma}$ ($\Sigma r^2 = 0.0146$). The water content of No. 80602 magma is calculated using water content of No. 80903 magma and of fractionated minerals. The water content of No. 80903 magma is calculated to be 6.8% by Burnham's method, and that of fractionated minerals is assumed to be 2% in amphibole, 4% in biotite, and 0% in other minerals. Thus, the water content of No. 80602 magma is calculated

to be 4.2% (Fig. 3). If water was introduced from the wall rocks into the crystallizing quartz monzodiorite magma, the initial water content of No. 80602 magma may be lower than the value obtained here. Therefore, water content of less differentiated magma seems to be lower in the Pl series than in the Am series. This estimation is consistent with the previous investigation by Tsuchiya (1985a).

Conclusions

1. Cl content of apatite in the Pl series rocks of the Matsumae plutonic complex is rather high and slightly increases within a range of low D.I., and abruptly decreases at about D.I.=45. On the other hand, Cl content of apatite in the Am series rocks is generally low and slightly decreases with differentiation.
2. F content of apatite shows no significant difference between the two series, and slightly increases with differentiation.
3. Such behavior of Cl and F in apatite

can be explained in terms of the hypothesis that more Cl was incorporated into the exsolved vapor relative to F, and removed from the magma.

4. Vapor exsolution may not play a significant role on the oxidation of magma.

5. The abrupt decrease in Cl without decrease in F content of apatite during the course of crystallization of magma is a useful indicator of vapor saturation, which provides valuable information about water content of magma. Estimated water content of the Matsuura plutonic magma is consistent with the previous results of petrological investigation.

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西南北海道松前深成岩類の燐灰石中の Cl および F 含有量

——揮発性成分飽和の指標——

土 谷 信 高

松前深成岩類の燐灰石中の Cl と F を、EPMA で定量した。松前深成岩類は、Am・Pl の 2 系列に分けられ、Am 系列はより水に富む条件下で固結したと考えられる。燐灰石中の Cl は、Am 系列では低く、Pl 系列では未分化な岩石中で高く D.I.=45 付近で急減している。また、燐灰石中の F は、両系列で差はみられず分化とともにわずかに増加している。

このような Cl と F の挙動は、マグマが揮発性成分に飽和して気相を分離したとき、Cl は F よりもより気相中に濃集しやすいためマグマから取り去られた、と考えることにより説明できる。すなわち、Am 系列のマグマは、Pl 系列よりも結晶作用のより早期に揮発性成分に飽和したのであろう。このように、F が減少せず Cl が急減する現象は、マグマが揮発性成分に飽和したことの良い指標となり、マグマ中の水の含有量の推定にもきわめて有効であらう。