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Fluid components in accessory minerals of Pan-African granitoids in the Sør Rondane Mountains, East Antarctica*

LI Zi-long†[1,5], CHEN Han-lin[1], YANG Shu-feng[1], TAINOSHO Yoshiaki[2],
SHIRAISHI Kazuyuki[3], OWADA Masaaki[4]
(*[1]Department of Earth Science, College of Science, Zhejiang University, Hangzhou 310027, China*)
(*[2]Faculty of Human Development, Kobe University, Nada, Kobe 657-8507, Japan*)
(*[3]National Institute of Polar Research, Kaga 1-chome, Itabashi-ku, Tokyo 173-8515, Japan*)
(*[4]Department of Earth Sciences, University of Yamaguchi, Yamaguchi 753-8512, Japan*)
(*[5]Department of Earth and Planetary Science, Harvard University, Cambridge, MA 02138, USA*)
†E-mail: zilongli@zju.edu.cn

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Abstract: Fluids (fluorine, chlorine, and OH) in accessory minerals (apatite, titanite and allanite) of Pan-African granitoids (Group-I granitoids, Group-II granitoids and Mefjell Plutonic Complex) from the Sør Rondane Mountains, East Antarctica were precisely measured by an electronic microprobe analyzer in this study. Apatites in the granites have commonly high fluorine contents. However, fluorine contents from the Group-I, Group-II granitoids and Mefjell Plutonic Complex (MPC) are of important variation, which F contents (3.21~7.20 wt%) in apatite from the Group-II granitoids are much higher than those from the Group-I granitoids (1.22~3.60 wt%) and the MPC (3.21~4.11 wt%). Titanite in the MPC has a low fluorine content (0.23~0.50 wt%), being less than those in the Group-I granitoids (2.28 wt%) and Group-II granitoids (1.85~2.78 wt%). Fluorine in allanite in the Group-II granitoids seems to have much lower contents than those from the Group-I granitoids and the MPC. Higher fluorine contents in the titanite from the Group-II granitoids may be mainly controlled by late-magmatic fluid-rock interaction processes associated with melt, but may not be indicative of original magma contents based on its petrographic feature. Due to very lower chlorine contents from all of accessory minerals, the authors suggest that titanite and apatite with higher fluorine contents in the Group-II granitoids have much lower H_2O (OH) contents compared with those in the Group-I granitoids according to the partition among (F, Cl, OH). Fluorine contents in whole-rock samples show a variation from the higher in the Group-I granitoids to the lower in the Group-II granitoids and the MPC, which are consistent with the changes of those from the biotite and hornblende as well as fluorite occurred in the Group-I granitoids reported previously. Based on the above study of fluorine in accessory minerals and combined with the previous fluorine contents from biotites and hornblendes, the authors suggest that apatites and titanites with higher F contents in the Group-II granitoids and the MPC may not be an indicator of higher fluorine contents in whole-rock, which reflect fluorine contents in magma sources and/or late-thermal activity. Higher fluorine contents in apatite, titanite and allanite may be an additional evidence of A-type affinity.

Key words: Fluorine contents, Accessory minerals, Pan-African granitoids, Sør Rondane Mountains, Antarctica
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INTRODUCTION

Recent advances in crystal spectrometer design and operation allow for routine fluorine and chlorine analysis by microprobe, although long counting periods are required (Li *et al.*, 2003a). Fluorine in biotite can be used as an indicator of magmatic and fluid compositional changes during late-magmatic fluid-rock interaction processes associated with melt (Markl and Piazolo, 1998; Selby and Nesbitt, 2000). High fluorine content enhances the stability of biotite

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at high temperatures (Skjerlie and Johnston, 1993; Dooley and Patiño Douce, 1996; Bose *et al*., 2005). Loiselle and Wones (1979) and Collins *et al*.(1982) argued that the typical F content of biotite in A-type granites indicates a relatively high HF/H_2O ratio in the magma and that A-type granites form at low H_2O fugacity, low to moderate oxygen fugacity and high temperatures. Recently King *et al*.(1997) and Rajesh (2000) reported low F contents in biotite from aluminous A-type granite. However, fluorine contents in accessory minerals from Pan-African granitoids with A-type affinity are not well studied in the world. The Sør Rondane Mountains are mainly composed of the high-grade metamorphic rocks at the Late Proterozoic and Pan-African plutonic rocks. Li *et al*.(2003b) divided Pan-African granitoids in the Sør Rondane Mountains into the Group-I granites (Dufek and Lunckeryggen granites), Group-II granites (Austkampane, Pingvinane, Rogerstoppane, and Vikinghøgda granites), MPC and Lunckeryggen Syenitic Complex based on field occurrence, petrological, and geochemical study and isotopic data. Therefore, in this paper, we present fluorine contents in accessory minerals from the Group-I granitoids, the Group-II granitoids and the MPC and use these data as well as those data from biotite and hornblende reported previously to explain the change of fluorine contents in accessory minerals and provide a further information of A-type granites.

REGIONAL GEOLOGY

The Sør Rondane Mountains (22°E to 28°E, 71.5°S to 72.5°S) in Dronning Maud Land, East Antarctica (Fig.1) mainly consist of Late Proterozoic green-schist- to granulite-facies metamorphic rocks and Late Proterozoic to Early Paleozoic plutonic rocks with minor mafic dykes (Takahashi *et al*., 1990; Shiraishi *et al*., 1991; Shiraishi and Kagami, 1992; Osanai *et al*., 1992; Tainosho *et al*., 1992; 1993; Arakawa *et al*., 1994; Ikeda and Shiraishi, 1998; Li *et al*.,

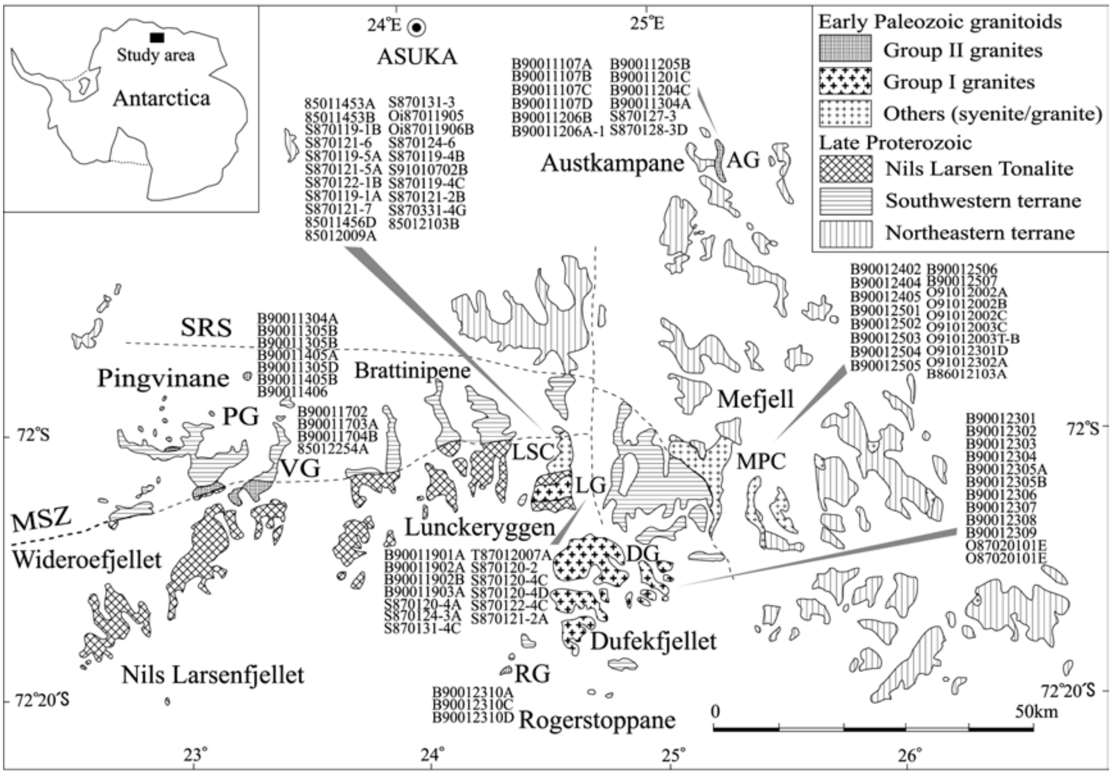

Fig.1 Simplified geological map of the Sør Rondane Mountains (modified from Shiraishi *et al.*, 1997) showing sample locality. The Group-I granites are: DG=Dufek granites and LG=Lunckeryggen granites. The Group-II granites are: AG=Austkampane granites, PG=Pingvinane granites, VG=Vikinghøgda granites and RG=Rogerstoppane granites. MPC=Mefjell Plutonic Complex; LSC=Lunckeryggen Syenitic Complex; MSZ=Main Shear Zone; SRS=Sør Rondane Suture Zone

2003a; 2003b; 2005; 2006). The region can be divided into two terranes on the basis of metamorphic grade: an amphibolite- to granulite-facies northeastern terrane and an epidote amphibolite- to greenschist-facies southwestern terrane. The Sør Rondane Suture Zone separates the two terranes, and the Main Shear Zone cuts the southwestern terrane (Osanai *et al.*, 1996).

Two episodes of 950 Ma and 500~620 Ma plutonisms were divided by the whole-rock Rb-Sr isochronal, Sm-Nd and SHRIMP U-Pb zircon methods (Takahashi *et al.*, 1990; Tainosho *et al.*, 1992; Shiraishi and Kagami, 1992; Ikeda and Shiraishi, 1998; Li *et al.*, 2005; 2006). Pan-African granitoids (500~620 Ma) are widely distributed in the Sør Rondane Mountains, representing a prominent Pan-African magmatic event. They are subdivided into the Group-I granites (Dufek and Lunckeryggen granites), Group-II granites (Austkampane, Pingvinane, Rogerstoppane, and Vikinghøgda granites), MPC and Lunckeryggen Syenitic Complex. Pan-African granitoids are composed mainly of granitic and syenitic rocks and are products of the regional mylonitization represented by the Main Shear Zone except for migmatic and foliated granites (Sakiyama *et al.*, 1988).

PETROGRAPHY AND GEOCHEMISTRY

Group-I granitoids: The Dufek granite outcropped in an area of approximately 10×10 km^2 in and around the Dufekfellet. It includes many tonalite xenolith in the north and huge blocks of the gneisses especially in the south. This granite is unfoliated and composed mainly of medium-grained biotite granite with fine-grained biotite granite and medium-grained biotite granodiorite. The medium-grained biotite granite is composed of plagioclase, K-feldspar, quartz and biotite with accessory titanite, apatite, zircon, muscovite and opaque minerals. The Lunckeryggen granite is exposed in an area of 6×6 km^2 in center of Lunckeryggen and associates with syenite. The granite intrudes into the Nils Larsen tonalite and quartz syenite of the Lunckeryggen syenitic complex and has angular xenoliths of these rocks. It is composed of a stock of coarse-grained granite and many dikes of fine-grained granite, and these rocks are composed of quartz, K-feldspar, plagioclase and biotite with or without hornblende, but with titanite, apatite, zircon, magnetite and occasionally fluorite (Table 1, Fig.2, and Li *et al.*, 2003b).

Group-II granitoids: The Austkampane granite is exposed in an area 1×3 km^2 in the eastern side of Austkampane. Mafic minerals of the granite show a parallel arrangement defining a foliation. Petrographically it has a coarse-grained granodiorite to granite. It consists of quartz, plagioclase, K-feldspar, hornblende and biotite with minor zircon, apatite, titanite and ilmenite. The Pingvinane granite occurs as stock and intrudes gneisses and includes xenolith of host gneisses. It is massive and equigranular coarse-grained biotite hornblende granite and is composed of K-feldspar, quartz, plagioclase, hornblende and biotite with accessory titanite, apatite, zircon and opaque minerals. The Rogerstoppane granite occurs at the southern end of Rogerstoppane and consists mainly of plagioclase, quartz, K-feldspar, biotite and hornblende with accessory allanite, epidote, zircon, apatite, titanite and opaque minerals with or without garnet. The Vikinghøgda granite is mainly

Table 1 Mineral assemblages of the Pan-African granitoids

Types		Kfs	Pl	Qtz	Hbl	Bt	Cpx	Aln	Ttn	Ap	Zrn	Grt	Mag	Ilm	Ep	Ms	Flt
Group-I granitoid	LG	++	++	++	±	+		+	+	+	+		+				+
	DG	++	++	++		+		+	+	+	+		+			±	±
Group-II granitoid	AG	++	++	++	±	+		±	+	+	+	+	+			+	
	PG	++	++	++	+	+	+	±	+	+	+		+				
	VG	++	++	++		+			+	+	+		+			+	
	RG	++	++	++	±	+		±	+	+	+	+	+		+	+	
MPC	Granite	++	++	++		+			+	+	+		+				
	Syenite	++	++	+	+	+	+	+		+	+		+	+		±	

Kfs: K-feldspar; Pl: plagioclase; Qtz: quartz; Hbl: hornblende; Bt: biotite; Cpx: clinopyroxene; Aln: allanite; Ttn: titanite; Ap: apatite; Zrn: zircon; Grt: garnet; Mag: magnetite; Ilm: ilmenite; Ep: epidote; Ms: muscovite; Flt: fluorite; ++: major mineral; +: minor or accessory mineral; ±: occasional occurring mineral

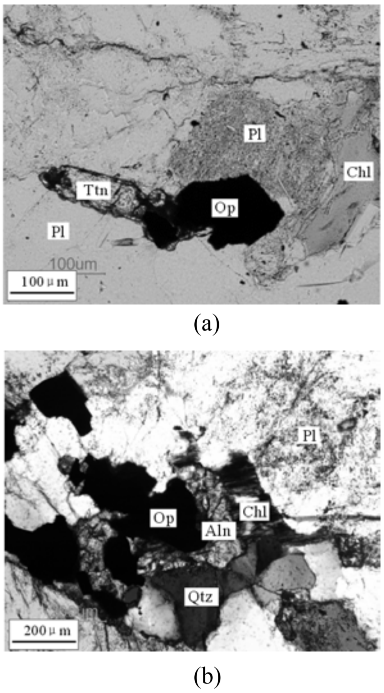

Fig.2 Photomicrography of the Lunckeryggen granite (Sample No. B90011902B), one of the Group-I granite. (a) Contact relationship of titanite, plagioclase, K-feldspar, quartz, chlorite and opaque mineral. Alkali granite. Single polars; (b) Mineral assemblage of allanite, plagioclase, K-feldspar, quartz and chlorite. Crossed polars.
Aln: allanite; Ttn: titanite; Pl: plagioclase; Op: opaque mineral; Chl: chlorite; Qtz: quartz

composed of quartz, plagioclase, K-feldspar, biotite and muscovite with accessory titanite, apatite, zircon and opaque oxides.

The MPC is exposed in an area of 7×10 km^2 in the eastern part of Mefjell. It consists of the syenite and granite and varies from granite composition in the northeast to an alkaline composition in the southwest. The syenite is medium- to coarse-grained, light gray in color and consists of K-feldspar, plagioclase, biotite, hornblende, Fe-rich clinopyroxene and quartz with minor apatite, zircon and iron oxide with or without Fe-rich olivine.

Geochemically, the Pan-African granitoids are granitic to syenitic in composition, and show alkaline affinity correlated to A-type granites worldwide based on major and trace element geochemistry (Li *et al.*, 2003b). These granitoids are characterized by high K_2O+Na_2O (7~13 wt%) and K_2O/Na_2O (1~2), low to intermediate Mg# (Mg/(Mg+Fe)), wide ranges of SiO_2 (45~78 wt%), Sr (20×10^{-6}~6500×10^{-6}) and Ba (40×10^{-6}~13000×10^{-6}), and having Nb and Ti depletion in the primitive mantle normalized diagram. The Group-I granitoids have higher Sr and F contents, higher Sr/Ba ratios, and lower Ga/Al ratios than the Group-II granitoids (Li *et al.*, 2003a). The syenite and granite of the MPC are alkaline, high in FeO/(FeO+MgO), Zr, Ba and Ga, and low in MgO, Rb, Y and Nb. The syenite has high LREE (light rare earth elements) concentration with positive Eu anomalies, whereas the granite has high LREE/HREE (heavy rare earth elements) ratios with negative Eu anomalies in the chondrite normalized REE diagram (Li *et al.*, 2005).

SAMPLES, METHOD AND RESULTS

Samples

The samples are those of the polished thin sections used to do electron microprobe analyses. These thin sections were petrographically described under a microscope, and some accessory minerals were circled with their description of property to use chemical analyses. Euhedral zircon and apatite are commonly enclosed in feldspar and/or mafic minerals of biotite and hornblende in the Group-I granitoids while zircon is included in feldspar and quartz in the Group-II granitoids. Titanites occurred in matrix with euhedral to subhedral. Darkly brown allanite crystals are euhedral to subhedral and have metamict cores and light-colored rims. Allanites and titanites are formed as the products of late-stage magmatic process (Fig.2).

Method and results

Chemical analyses of accessory minerals (apatite, titanite and allanite) were obtained using a JEOL-8900M electron probe microanalyzer at the Venture Business Laboratory of Kobe University, Japan. Element determinations were carried out using a beam size of 3 μm, an accelerating potential of 15 kV, a probe current of 12 nA. For fluorine and chlorine analyses, natural pure fluorite (CaF_2) and a granular aggregate of halite crystal (NaCl) were adopted as standards. The interference of Fe-Lα line with F-Kα line was removed by calculation. Counting times of the peak and background for F and Cl were set as 100 s and 50 s respectively. Matrix effects during analytical procedure were corrected using the ZAF software provided by JEOL. Accuracy of the fluorine and chlorine analyses examined by measurements of the fluorite and NaCl standards is within the standard error of the analyses of c. 1 wt% (2 sigma). Detection limits of F and Cl are usually 0.01 wt% (3 sigma). Each microprobe analysis of accessory minerals is an average of three spot analyses (Li *et al.*, 2003a). Concentrations of F and Cl in apatite, titanite, allanite, biotite and hornblende are listed in Table 2.

Apatite and titanite grains in the Group-II granitoids have much higher F contents, whereas allanite have lower F contents compared to those in the other granitoids (Table 2). Though apatites from the granitoids measured have commonly high fluorine contents, but apatites from the Group-II granitoids have higher fluorine contents, ranging from 3.21~6.93 wt%, compared to those from the Group-I granitoids (1.00~3.60 wt%) and MPC (3.20~3.95 wt%) (Table 2). Titanite in the MPC has a low fluorine content (0.35 wt%), being less than those in the Group-I granitoids (0.80~1.60 wt%) and the Group-II granitoids (2.44 wt%) (Table 2). Allanite in the Group-II granitoids has fluorine contents of 0.32 wt%, being much lower contents than those from the Group-I granitoids (1.70~2.22 wt%) and MPC (0.23~0.50 wt%). In allanite from Group-II granitoids, chlorine contents of less than 0.05 wt% are higher than those of <0.02 wt% in that of Group-I granitoids. However, apatite and titanite grains from all samples of granitoids have very low chlorine of less than 0.02 wt% or no-detection.

DISCUSSION AND SUMMARY

The different fluid components in the accessory minerals from three types of granitoids (Group-I granitoids, Group-II granitoids and MPC) may be reflected by variable magmatic processes of Pan-African granitoids. As Li *et al.*(2003b) argued that a mantle-derived, hot basic magma (altered composition) in lower crustal or upper mantle level, during ascending, produced the Group-I granitic melt by a process of fractional crystallization with minor assimilation or mixing. The Group-II granites may be derived from the same hot magma by a process of assimilation with crustal rocks and consequently fractional crystallization in higher crustal levels (ACF model). The MPC seems to be derived from a heterogenetic magma source and syenitic rocks in it have a unique source (iron-enriched) judging from their chemical data. Since higher F in apatite is usually as melt component and shows an igneous genetic, not fluid phase according to F vs Cl diagram as pointed by Owada and Morifuku (2001), whereas allanite often occurs in the metamict state because destruction of the crystalline structure metamictization lowers the stability and such allanite becomes more susceptible to alteration (Deer *et al.*, 1996). Therefore, the increases of fluorine contents in the apatites from the Group-I granitoids to Group-II granitoids, in which the apatites are regarded as an early-stage products in the formation of granitoids due to their state of occurrence, may indicate that magmatic fluid (fluorine) components increase during magma going up from the deep level towards the shallow level of crust.

Table 2 Chemical composition of fluorine and chlorine contents of accessory and mafic minerals from Pan-African granitoids

Types		F (wt%)						Cl (wt%)					
		Ap	Ttn	Aln	Zrn	Hbl	Bt	Ap	Ttn	Aln	Zrn	Hbl	Bt
Group-I granitoid	LG	3.40~3.60	0.8~1.6	1.7~2.22	0.12		1.50~1.70	n.d.	0.01	0.01	0.02		0.01~0.06
	DG	1.00~1.60		0.80~2.31	n.d.	1.15~1.23	1.86~2.32			0~0.02	0~0.01	0.28~0.32	0.03~0.05
Group-II granitoid	AG				0.05~0.06	0.29	0.5				n.d.	0.36	0.3
	PG	3.21~6.93			0.06~0.18	0.25~0.43	0.50~0.78	0~0.02			n.d.	0.31~0.41	0.04-~0.41
	VG						0.40~0.55						0.05
	RG	5.32	2.44	0.32	0.07	0.45	0.8	0.003	0.017	0.05	0.022	0.21	0.15
MPC			3.20~3.95	0.35	0.23~0.50	0.09~0.05	0~0.01	0~0.01	0~0.02	0.004	0.03	0.003	0.11~0.26

Note: Hornblende data of MPC is from syenite, not granite; Fluorine and chlorine data of hornblende and biotite were from (Li *et al.*, 2003a); n.d.: no-detection

Higher fluorine contents in titanite from the Group-II granitoids may be mainly controlled by late-magmatic fluid-rock interaction processes associated with melt, but may not be indicative of original magma contents. Fluorine in allanite in the Group-II granitoids having much lower contents than those from the Group-I granitoids and MPC, cannot explain the variations of the late-magmatic thermal effect.

The higher fluorine contents in apatites from the Group-II granitoids may not explain the changes of fluorine contents in whole-rock. Because fluorine contents in whole-rock from the Group-I granitoids, Group-II granitoids and the MPC are 100×10^{-6}~2600×10^{-6}, 50×10^{-6}~1500×10^{-6} and 100×10^{-6}~400×10^{-6} respectively (Li *et al.*, 2003a), this means that the Group-I granitoids have much higher fluorine contents than those of the Group-II granitoids and the MPC. Through CIPW calculation, in general, these granitoids have relatively higher components in apatite and titanite, and lower in allanite, therefore, apatite and titanite should control mainly the whole-rock fluorine contents than allanite among accessory minerals analyzed. However, as described above, the Group-I granitoids have higher modal components of biotites and hornblendes (high fluorine contents) as well as fluorite occurred, thus, the higher fluorine contents in whole-rock compositions of the Group-I granitoids than those in the other granitoids (Li *et al.*, 2003a) may be mainly attributed to the existence of biotite and hornblende grains and fluorite.

A-type granitic magmas typically contain higher amounts of fluorine and chlorine than I-type granitic magmas (Loiselle and Wones, 1979; Collins *et al.* 1982). As Collins *et al.*(1982) suggested, if fluorine is relatively low in the melt, early amphibole crystallization may not occur and anorthite-rich plagioclase is the dominant crystallizing phase. Fluorine concentration in the melt is therefore considered to be critical in determining fractionation trends in A-type magmas. Consequently, the high fluorine contents in accessory minerals, particularly apatites and titanites from the Pan-African granitoids in the study area may be regarded as an additional evidence of A-type granites.

CONCLUSION

(1) Fluorine contents as main fluid components in apatite from the Group-II granitoids (3.21~7.20 wt%) is much higher than those from the Group-I granitoids (1.22~3.60 wt%) and the MPC (3.21~4.11 wt%). Titanite in the MPC has a low fluorine content (0.23~0.50 wt%), being less than those in the Group-I granitoids (2.28 wt%) and the Group-II granitoids (1.85~2.78 wt%); and fluorine in allanite has much lower contents in the Group-II granitoids than those from the Group-I granitoids and the MPC. Titanite and apatite with higher fluorine contents in the Group-II granitoids have much lower H_2O (OH) contents compared with those in the Group-I granitoids.

(2) Apatites and titanite with higher F in the Group-II granitoids and MPC may not be an indicator of higher fluorine contents in whole-rock based on fluorine contents in whole-rock samples from the Group-I granitoids, the Group-II granitoids and the MPC.

(3) Higher fluid components (fluorine contents) in apatites and titanites from the Group-I granitoids, Group-II granitoids and the MPC may be an additional implication of the Pan-African granitoids with A-type affinity.

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