OKAYAMA University
Earth Science Reports,
Vol. 10, No.1, 1-10, (2003)

Zinc Contents of Mafic Minerals in Granitic Rocks, with Special Reference to Ore Chemistry

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In order to know the reason why the Cu/Zn-Pb ratios of skarn-type deposits related with the ilmenite-series granitic rocks are markedly higher than those related with the magnetite-series, comparative mineralogical studies were carried out for the two types of granitic rocks in the Chugoku district. An attention was focused on the behavior of Zn during the crystallization of granitic magma, because Zn contents of mafic minerals can be obtained by non-destructive electron microprobe analyses.

Microscopic observation indicates that no sphalerite occurs in the magnetite-series granitic rocks, which implies that the magmas corresponding to the granitic rocks were undersaturated in respect to ZnS. Electron microprobe analyses revealed that the Fe/(Fe+Mg) ratios of mafic minerals such as biotite and hornblende in the ilmenite-series granitic rocks are remarkably higher than those in the magnetite-series, and the Zn contents are positively correlated with the Fe/(Fe+Mg) ratios in the ilmenite-series granitic rocks. Also the Zn/Fe ratios seem to be slightly higher in the ilmenite-series granitic rocks. In contrast, the correlation between Zn contents and Fe/(Fe+Mg) ratios is ambiguous in magnetite-series granitic rocks.

With a progress of crystallization differentiation, therefore, Zn may be removed more effectively in the ilmenite-series granitic magma, and the contents may be growing scarce in the fractionated ilmenite-series granitic magma. On the other hand, magnetite-series granitic magma probably increases the Zn-contents in the advanced stage of the crystallization. The hydrothermal fluids genetically related to the fractionated magnetite-series granitic magma are likely to be enriched in Zn, being favorable for the formation of Zn-rich deposits.

Keywords: Magnetite-series, Ilmenite-series, Granitic rocks, Zn contents, Mafic minerals, Ore deposit.

I. Introduction

In the Inner Zone of Southwest Japan, magnetite-series granitic rocks, frequently porphyritic in texture, are dominantly distributed in the San-in belt, while equigranular ilmenite-series granitic rocks are common in the Sanyo belt. The former carries porphyry copper-molybdenum deposits, and the latter accompanies greisen-type tin-wolframite deposits (Ishihara, 1977). Shimazaki (1975) assessed the regional characteristics of mineralization in Japanese pyrometasomatic deposits, and indicated that relatively Cu-rich deposits tend to be associated with plutonic igneous activity, while Zn-Pb-rich ones are associated with hypabyssal to effusive activity. The Cu-rich and Zn-Pb-rich belts are recognized in the Inner Zone of Southwest Japan, and the two belts probably correspond to the Sanyo and San-in belts. Therefore, ilmenite-series and magnetite-series granitic rocks are likely to be genetically related with the Cu-rich and Pb-Zn-rich mineralizations, respectively. The parallelism strongly suggests that fractional crystallization is intimately related with the enrichment and depletion of Cu, Zn and Pb in the melt. Of these elements, Zn contents of mafic minerals can be obtained by non-destructive method on the electron-probe microanalyzer (EPMA) (Nakano, 1993).

In order to know the behavior of Zn during the crystallization of granitic magma, Sakai (1998) obtained by EPMA analyses the Zn contents of mafic minerals in granitic rocks at Kurashiki, which host several greisen-type tungsten deposits such as those at Miyoshi and Mizuwakare, and showed that the Zn contents of mafic minerals as biotite and hornblende from the ilmenite-series granitic rocks are generally higher than those from the magnetite-series granitic rocks, and that the Zn contents are positively correlated with the Fe contents. Kitamura (1999) carried out similar studies for magnetite-series granitic rocks in the Yubara area, northern Okayama Prefecture, and pointed out that Zn contents are positively correlated with Mn contents in biotite, hornblende and ilmenite. The results summarized above suggest that the magmas brought about the ilmenite-series granitic rocks become poorer in Zn contents with a progress of crystallization, compared with those of the magnetite-series granitic magma. However, further data are needed to come to a firm conclusion about the behavior of Zn during the crystallization of granitic magma. Therefore comparative studies on the behavior of Zn during the crystallization of granitic magmas are intended in the present study between ilmenite-series and magnetite-series granitic bodies distributed in the eastern Chugoku district.

II. Geology and Samples

Rock samples provided for analyses were taken from the granitic masses shown in Figure 1. They include 13 samples of magnetite-series granitic rocks from the Nichinan and Yubara masses and 19 samples of ilmenite-series granitic rocks taken from the Nariwa, Nibori and Mannari masses (Fig. 1). According to Murayama et al. (1973) and Hattori (1978), granitic rocks near the Nichinan Town are composed of three discrete masses, which are named the Kami-iwami, Yokota and Sangunsan in this study. The plutonic rocks in the Yubara area consist of the Inatsu, Yubara South, Yubara North and Unanji masses (Fig. 2). Metallic mineral deposits are associated with some of these granitic rocks. The Ohkura vein-type Pb-Zn-Cu deposit is associated with the Kami-iwami mass. The skarn-type Sanpo Cu-Zn-Fe deposit and Sano Fe-Pb-Cu deposit are with the Nariwa and Nibori masses, respectively.

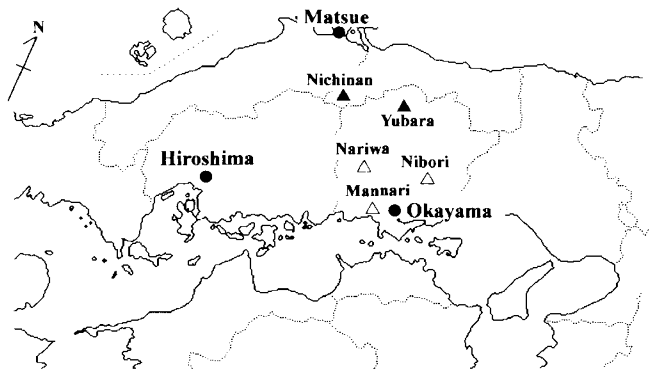

Fig. 1. Location of the sampled granitic rocks. Open and solid triangles indicate ilmenite-series and magnetite-series granitic rocks, respectively

The K-Ar ages are 69±6Ma for Kami-iwami (Hattori and Shibata, 1974), about 50Ma for Yokota (Kawano and Ueda, 1967), 56.9±1.7Ma for Sangunsan (Kitagawa et al., 1988), 79.9±2.8Ma for Nibori (Shibata, 1979) and about 86Ma for Mannari (Kawano and Ueda, 1966). Five stages are discriminated from mutual geological relations for the intrusion of plutonic rocks in the Yubara area (Sasada, 1978). The Inatsu mass is the earliest in intrusion and is assigned to stage I. The Yubara South mass is to stage II. The Yubara North and Unanji masses are to stage III. The age data of these masses determined by the Rb-Sr method are, 85.2±1.7Ma for Inatsu, 70.0±0.02Ma for Yubara South, 65.4±3.0Ma for Yubara North, and 69.0±

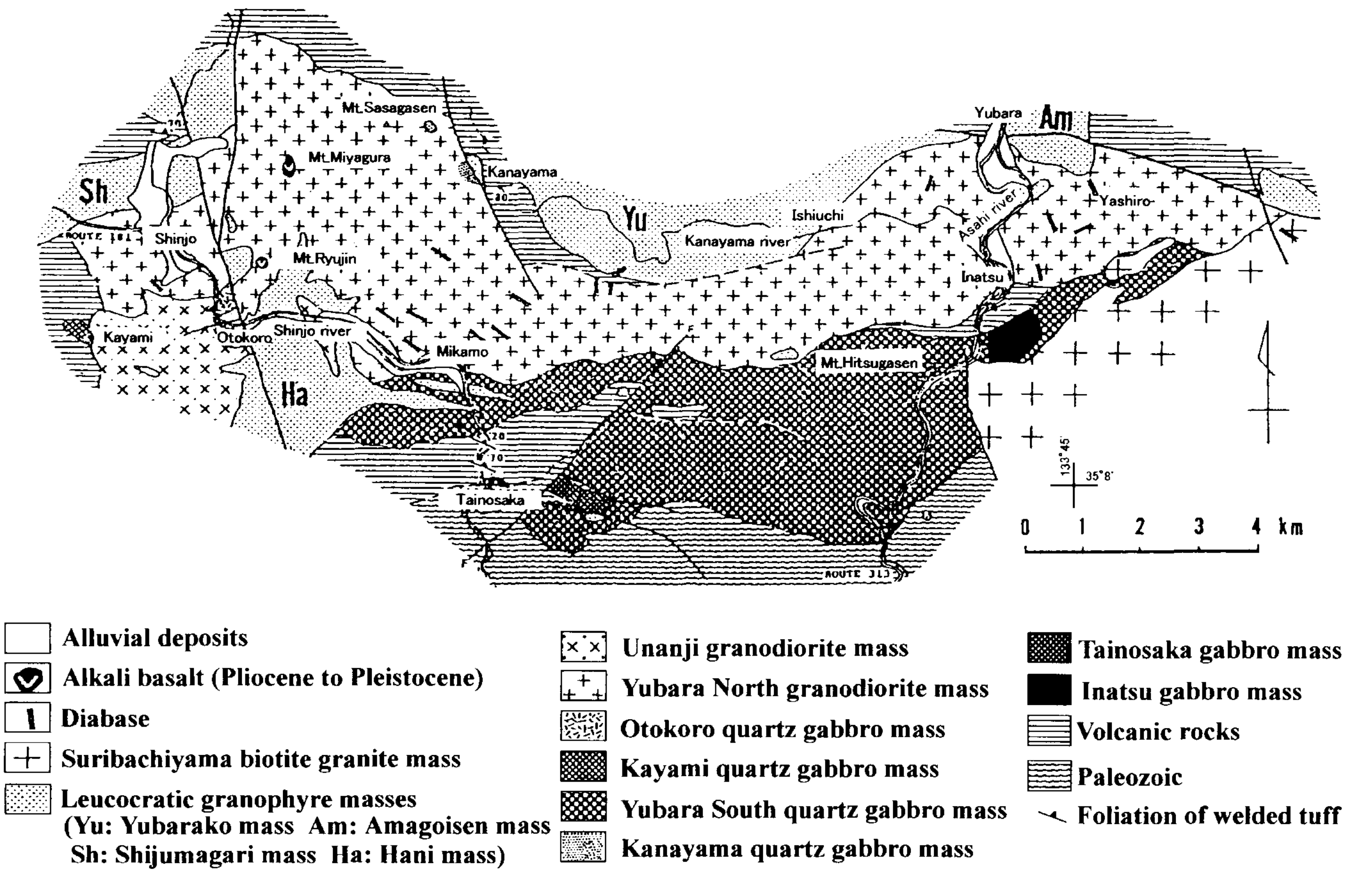

Fig. 2. Geological map of the Yubara area (partly modified from Sasada, 1978)

Location	Number	pl	Kf	qz	bt	am	cpx	ch	ep	mt	il	sp	py	cp	m.s.	△value
Kami-iwami	00060901	○	△	△	+	+	-	+	+	+	+	-	-	+	6.6	-
Yokota	00060902a	◎	△	△	+	+	-	+	+	+	+	-	+	+	5.8	<0.2
	00060902b	◎	△	○	+	+	-	+	+	+	+	-	+	-	6.8	<0.2
Sangunsan	00060903	△	○	◎	+	-	-	-	-	+	+	-	-	-	4.2	<0.2
	00060904	○	○	△	+	-	-	+	+	+	+	-	-	-	5.0	0.89
	00060905	△	△	○	+	-	-	+	-	+	+	-	-	-	2.9	<0.2
Inatsu	AK9801	◎	-	-	+	△	○	+	-	+	+	-	-	-	5.0	-
Yubara North	AK9802	◎	△	△	+	△	-	+	+	+	+	-	-	-	10	-
	AK9803	◎	+	△	+	△	-	+	+	+	+	-	+	+	20	-
Yobara South	AK9804	◎	+	△	+	△	-	+	+	+	+	-	-	-	10	-
	AK9805	◎	+	△	+	△	-	+	+	+	+	-	+	+	16	-
Unanji	AK9806	◎	△	○	+	+	+	+	-	+	-	-	+	+	22	-
	AK9807	◎	△	○	+	+	-	+	+	+	+	-	+	+	19	-
Mannari	111207	◎	△	△	+	+	-	+	+	+	+	-	-	-	0.9	0.37
	111208a	○	○	△	+	+	-	+	-	-	+	+	-	+	1.0	0.5
	111208b	○	△	◎	+	+	-	+	+	-	+	-	-	-	1.1	0.5
Nariwa	01072701	○	△	○	+	-	-	+	-	-	-	-	-	-	0.06	<0.2
	01072702	△	△	◎	+	-	-	+	-	-	+	-	-	-	0.03	<0.2
	01072703	△	○	○	+	-	-	+	-	-	+	-	-	-	0.05	<0.2
	01072704	○	◎	△	+	-	-	+	+	-	+	-	-	-	0.06	<0.2
	01072705	○	△	◎	+	-	-	+	+	+	+	-	-	-	0.05	<0.2
	01072706	△	○	△	+	-	-	+	+	-	+	-	-	-	0.05	<0.2
Nibori	02072701	○	△	○	+	+	-	+	-	+	+	-	+	-	0.10	<0.2
	02072702	◎	△	○	+	+	-	+	+	+	+	-	+	-	0.13	<0.2
	02072704	○	△	○	+	+	-	+	-	+	+	+	-	+	0.15	<0.2
	02111801	○	△	○	△	+	-	+	+	+	+	-	-	-	0.36	<0.2
	02111802	○	○	△	+	+	-	+	-	-	+	-	+	-	0.13	-
	02111803	○	△	△	+	+	-	+	+	-	+	-	+	-	0.12	0.59
	02111804	○	○	○	+	+	-	+	+	+	+	-	-	-	0.89	-
	02111805	○	△	△	+	+	-	+	+	+	+	-	-	-	0.43	-
	02111806	○	○	○	+	+	-	+	-	+	+	-	-	-	0.77	-

abundance	◎	○	△	+	—
	abundant	large	moderate	small	not observed

Fig. 3. Constituent minerals and their relative abundance of granitic rocks relevant to the present study. Abbreviations are pl: plagioclase, Kf: K-feldspar, qz: quartz, bt: biotite, am: amphibole, cpx: clinopyroxene, ch: chlorite, ep: epidote, mt: magnetite, il: ilmenite, sp: sphalerite, py: pyrite, cp: chalcopyrite and m.s.: magnetic susceptibility. Refer to the text on the Δ value.

2.2Ma for Unanji (Sudo et al., 1988).

III. Experimental technique

Petrographical studies in the present study mainly consist of microscopic observations and electron microprobe analyses. Biotite, hornblende, feldspars, ilmenite and magnetite were analyzed by using JXA-733 electronprobe microanalyzer, operating at an accelerating voltage of 15 kV and a specimen current was 20nA on Cu metal. An accelerating voltage of 20 kV was used for measurements of ZnKα because of the very weak intensities for samples. For ZnKα, measurements of the background were carefully carried out in particular. The X-ray intensity data were corrected according to Bence and Albee (1968). No correction procedures were applied to Zn except for the background correction according to Nakano (1968). Representative microprobe analyses of biotite and hornblende are shown in Tables 1 and 2. Zinc can not be detected in plagioclase, K-feldspar and quartz.

Triclinicity (Δ value) of K-feldspar was calculated from the X-ray powder diffraction data according to Goldsmith and Laves (1954). Analytical conditions were CuKα radiation, voltage 30kV, current 15mA and scan speed 2θ = 0.5°. The magnetic susceptibility of rocks was obtained by a Geofyzika Brno Kappameter KT-5.

IV. Petrography

Figure 3 shows the constituent minerals and their relative abundance of granitic rocks relevant to the present study. Magnetic susceptibility indicates that all the granitic rocks in the Nichinan and Yubara areas belong to the magnetite-series. The Nariwa, Nibori and Mannari masses

Table 1. Representative electron microprobe analyses of biotite

	magnetite-series							ilmenite-series			
No.	1	2	3	4	5	6	7	8	9	10	11
	060901	060901	060902a	060903	AK9802	AK9804	AK9807	072706	072701	111208b	111208b
weight %											
SiO_2	36.62	36.21	36.84	39.29	37.87	36.84	37.95	34.40	34.38	33.47	33.99
TiO_2	6.54	6.57	6.68	4.27	4.37	4.50	4.27	5.40	7.10	4.77	4.01
Al_2O_3	12.99	12.99	13.31	12.32	13.14	13.19	13.28	13.15	13.61	12.52	13.02
FeO*	23.34	23.07	17.27	13.27	20.07	20.31	17.45	31.18	27.98	33.54	33.36
MnO	0.32	0.33	0.99	0.94	0.27	0.21	0.71	0.44	0.50	0.78	0.73
MgO	9.41	9.30	12.39	16.90	11.94	11.87	13.58	3.86	5.03	2.68	3.05
CaO	0.01	0.00	0.00	0.03	0.00	0.00	0.00	0.03	0.01	0.00	0.01
Na_2O	0.06	0.06	0.06	0.15	0.09	0.15	0.11	0.05	0.21	0.05	0.06
K_2O	9.15	9.05	8.98	9.16	8.94	9.28	9.16	8.35	8.27	8.35	8.25
Total	98.44	97.58	96.52	96.33	96.69	96.35	96.51	96.86	97.09	96.16	96.48
ppm											
ZnO	250	253	555	320	372	309	663	401	605	926	855
number of atoms per formula unit (O=22)											
Si	5.527	5.512	5.521	5.767	5.701	5.601	5.672	5.493	5.391	5.482	5.522
Ti	0.742	0.752	0.753	0.471	0.495	0.515	0.480	0.649	0.837	0.588	0.490
Al	2.311	2.330	2.351	2.131	2.331	2.364	2.339	2.475	2.515	2.417	2.493
Fe	2.946	2.937	2.164	1.629	2.527	2.583	2.181	4.164	3.669	4.594	4.532
Mn	0.041	0.043	0.126	0.117	0.034	0.027	0.090	0.060	0.066	0.108	0.100
Mg	2.117	2.110	2.768	3.698	2.679	2.691	3.026	0.919	1.176	0.654	0.739
Ca	0.002	0.000	0.000	0.005	0.000	0.000	0.000	0.005	0.002	0.000	0.002
Na	0.018	0.018	0.017	0.043	0.026	0.044	0.032	0.015	0.064	0.016	0.019
K	1.762	1.757	1.717	1.715	1.717	1.800	1.747	1.701	1.654	1.745	1.710
Total	15.465	15.459	15.418	15.575	15.510	15.624	15.567	15.479	15.374	15.603	15.606
Fe/(Fe+Mg)	0.582	0.582	0.439	0.306	0.485	0.490	0.419	0.819	0.757	0.875	0.860
Mn/(Fe+Mg+Mn)	0.008	0.008	0.025	0.021	0.007	0.005	0.017	0.012	0.014	0.020	0.019
Zn/Fe ($\times 10^{-3}$)	0.779	0.797	2.239	1.660	1.349	1.111	2.691	0.935	1.566	1.988	1.849

* Total Fe as FeO. 1 and 2: Kami-iwami; 3: Yokota; 4: Sangunsan; 5: Yubara North; 6: Yubara South; 7: Unanji; 8:Nariwa; 9:Nibori; and 10 and 11: Mannari. Nos.1 and 2, and 10 and 11 indicate the composition of core and rim of grain, respectively.

have magnetic susceptibility corresponding to that of the ilmenite-series. Brief descriptions of these granitic rocks are given here. The subdivision of granitic rocks is due to Streckeisen (1973).

1. Magnetite-series granitic rocks

(1) Granitic masses in the Nichinan area

The Kami-iwami and Yokota masses contain hornblende and biotite as mafic minerals, and they are classified into hornblende-biotite granite and biotite-hornblende granite, respectively (Fig. 4-a). The Sangunsan samples include only biotite as a mafic mineral, thus the mass is classified into a biotite granite. All the granitic rock samples in the Nichinan area are found to have euhedral to subhedral plagioclase with a distinct compositional zoning and anhedral K-feldspar with a perthitic texture. K-feldspar in the Kami-iwami sample is characteristically associated with corroded quartz. Concerning the triclinicity of K-feldspar, the delta values are mostly smaller than 0.2 in this area, but the value is exceptionally high, 0.89, in a Sangunsan sample. Hornblendes in the Kami-iwami and Yokota samples show zonal structures under microscope. No zonal structures of biotite can be observed under microscope. For the Kami-iwami biotite, however, a chemical inhomogeneity is only slightly detected by back-scattered electron image of EPMA.

Table 2. Representative electron microprobe analyses of hornblende

No.	magnetite-series							ilmenite-series		
	1	2	3	4	5	6	7	8	9	10
	060901	060901	060902a	AK9801	AK9802	AK9805	AK9807	111801	111208b	111208b
weight %										
SiO_2	46.68	44.63	54.20	53.85	48.49	48.27	49.00	42.03	40.83	41.03
TiO_2	1.85	2.37	0.32	0.17	1.09	0.93	0.98	2.58	3.30	3.40
Al_2O_3	5.48	7.06	1.77	1.54	5.91	5.67	5.32	8.67	8.57	8.64
FeO*	18.18	18.99	9.83	12.72	16.32	15.94	13.52	24.66	28.00	27.91
MnO	0.48	0.45	1.43	0.23	0.50	0.33	1.08	0.57	0.69	0.69
MgO	11.38	10.67	17.72	16.81	13.11	13.79	15.41	6.15	3.95	3.87
CaO	11.51	11.35	11.41	12.12	11.15	11.27	10.61	10.31	10.03	10.01
Na_2O	1.23	1.47	0.00	0.14	1.34	1.25	1.10	1.78	2.14	2.20
K_2O	0.69	1.05	0.10	0.05	0.58	0.59	0.33	0.90	1.19	1.13
Total	97.48	98.04	96.78	97.63	98.49	98.04	97.35	97.65	98.70	98.88
ppm										
ZnO	246	119	284	130	74	267	392	486	530	403
number of atoms per formula unit (O=23)										
Si	7.046	6.765	7.784	7.754	7.136	7.126	7.191	6.581	6.466	6.476
Ti	0.210	0.270	0.035	0.018	0.121	0.103	0.108	0.304	0.393	0.404
Al	0.975	1.261	0.300	0.261	1.025	0.987	0.920	1.600	1.599	1.607
Fe	2.295	2.407	1.181	1.532	2.008	1.968	1.659	3.229	3.708	3.684
Mn	0.061	0.058	0.174	0.028	0.062	0.041	0.134	0.076	0.093	0.092
Mg	2.561	2.411	3.794	3.609	2.876	3.035	3.371	1.436	0.932	0.911
Ca	1.861	1.843	1.756	1.870	1.758	1.783	1.668	1.730	1.702	1.693
Na	0.360	0.432	0.000	0.039	0.382	0.358	0.313	0.540	0.657	0.673
K	0.133	0.203	0.018	0.009	0.109	0.111	0.062	0.180	0.240	0.228
Total	15.503	15.651	15.041	15.121	15.477	15.512	15.428	15.675	15.790	15.767
Fe/(Fe+Mg)	0.473	0.500	0.237	0.298	0.411	0.393	0.330	0.692	0.799	0.802
Mn/(Fe+Mg+Mn)	0.012	0.012	0.034	0.005	0.013	0.008	0.026	0.016	0.020	0.020
Zn/Fe ($\times 10^{-3}$)	0.972	0.451	1.857	0.704	0.324	1.210	1.348	1.420	1.362	1.039

* Total Fe as FeO. 1 and 2: Kami-iwami; 3: Yokota; 4: Inatsu; 5: Yubara North; 6: Yubara South; 7: Unanji; 8:Nibori; and 9 and 10: Mannari. Nos.1 and 2, and 9 and 10 indicate the composition of core and rim of grain, respectively.

Opaque minerals are composed of magnetite and ilmenite, with or without trace amounts of sulfide minerals as pyrite and chalcopyrite. Pyrrhotite occurs in small amounts in the Kami-iwami samples. These sulfides are granular in shape and occur in mafic minerals and magnetite. Magnetite is replaced by hematite along the crystallographically preferred planes and the grain margin, probably as a result of oxidation. Ilmenite occurs as discrete grains and also as composite grains with magnetite.

(2) Granitic rocks in the Yubara area

Modal analyses indicate that the Inatsu mass is a gabbro, the Yubara South and Yubara North masses are biotite-hornblende granodiorite and the Unanji mass is a hornblende-biotite granite (Fig. 4-a). Commonly, plagioclase is euhedral or subhedral in these plutonic rocks, and has a zonal structure under microscope. In the Inatsu mass, clinopyroxene and acicular actinolite are observed, and no quartz and K-feldspar occur. In the plutonic rocks other than the Inatsu mass, quartz and K-feldspar occur in variable amounts. The amounts increase in the order of Yubara South, Yubara North and Unanji (Fig. 4-a).

Magnetite and ilmenite occur as discrete and composite grains in biotite, hornblende and plagioclase and/or in interstices of these mineral grains. Ilmenite is also found as exsolution lamellae in magnetite. Small amounts of granular chalcopyrite and pyrite are included in magnetite of samples from Yubara South, Yubara North and Unanji.

2. Ilmenite-series granitic rocks

(1) Nariwa mass

The Nariwa mass is a biotite granite (Fig. 4-b). Plagioclase is euhedral or subhedral and strongly saussuritized in the core. K-feldspar is anhedral with a perthitic texture, and it is sometimes associated with corroded quartz. The delta values are small (<0.2).

Small amounts of ilmenite and trace amounts of magnetite occur in biotite as inclusions, and also in interstices of common minerals.

(2) Nibori mass

The Nibori mass is classified into a hornblende-biotite granite (Fig. 4-b). The euhedral plagioclase shows concentric growth zoning. K-feldspar is anhedral with a perthitic and a micrographic texture. Most of the delta values are small (<0.2), except for one sample with value of 0.59. Tabular hornblende crystal is pleochroic from brownish green to pale green.

Ilmenite and magnetite appear as discrete and composite grains in small and trace amounts, respectively. Ilmenite also occurs as exsolution lamellae in magnetite. Sulfides are rarely observed in trace amounts, which include granular pyrite and pyrrhotite in biotite and ilmenite, and also in their interstices. The composite grain of chalcopyrite and sphalerite is rarely included in plagioclase.

(3) Mannari mass

The Mannari hornblende-biotite granite (Fig. 4-b) is characterized by the presence of pinkish K-feldspar. Subhedral to euhedral plagioclase shows concentric zoning. K-feldspar is anhedral with a perthite intergrowth. The delta values range from 0.37 to 0.50. Back-scattered electron images show that hornblende is chemically homogeneous and no compositional zoning is observed.

Small amounts of ilmenite and trace amounts of magnetite occur as discrete grains. Ilmenite is also observed as exsolution lamellae in magnetite. Pyrite and sphalerite appear as a composite grain in allanite.

V. Results

1. biotite and hornblende

In ilmenite-series granitic rocks, the Fe/(Fe+Mg) atomic ratios of biotite and hornblende range from 0.6 to 0.9 and from 0.65 to 0.9, respectively (Fig. 5). In magnetite-series, the Fe/(Fe+Mg) ratios are from 0.3 to 0.6 for biotite, and from 0.2 to 0.5 for hornblende. The ZnO contents of biotite are from 375 to 1125ppm with an average of about 700ppm for the ilmenite-series granitic rocks, while they are from 97 to 750ppm and average about

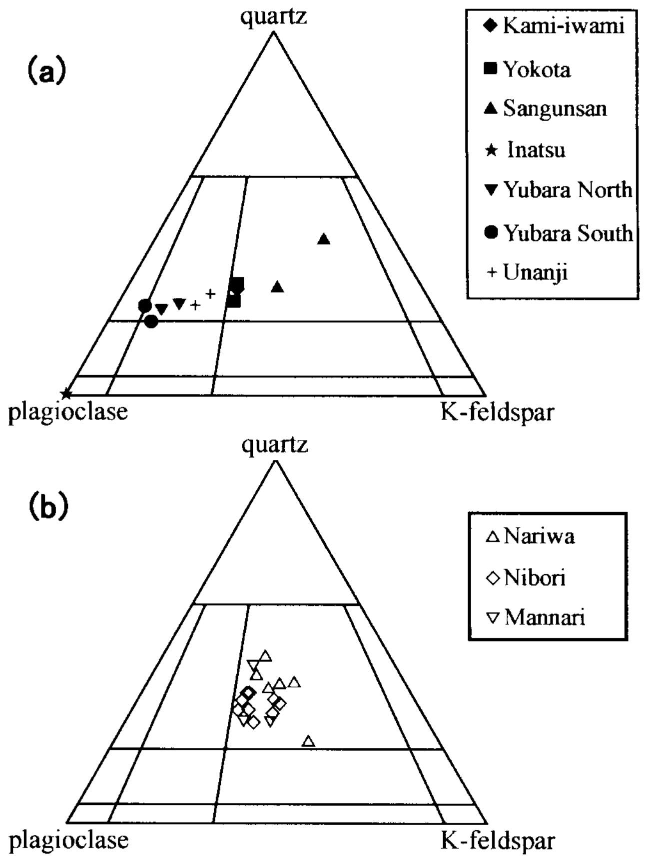

Fig. 4. Modal compositions of (a) magnetite-series and (b) ilmenite-series granitic rocks.

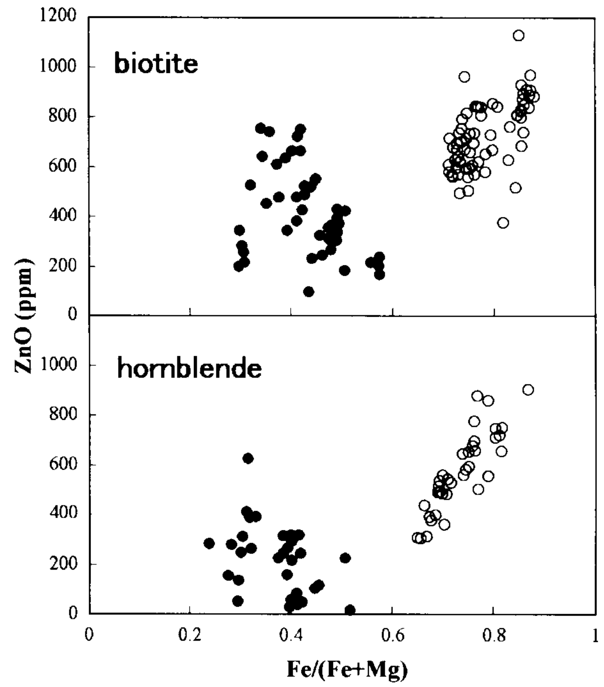

Fig. 5. Relationship between Fe/(Fe+Mg) atomic ratios and ZnO contents in mafic minerals. solid circles: magnetite-series granitic rocks; open circles: ilmenite-series granitic rocks.

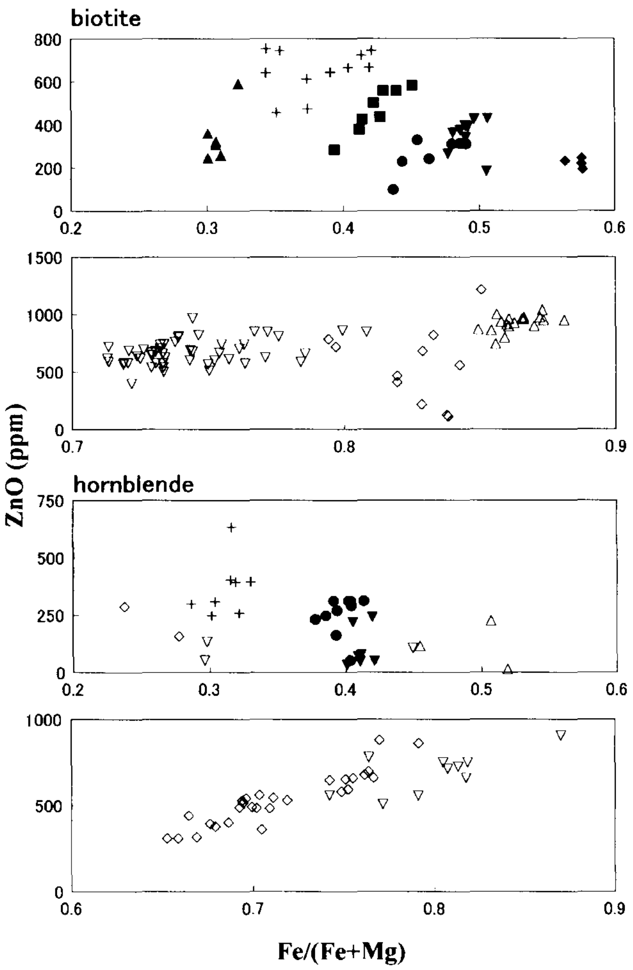

Fig. 6. Relationship between Fe/(Fe+Mg) atomic ratios and ZnO contents in mafic minerals. Symbols are the same as Fig.4.

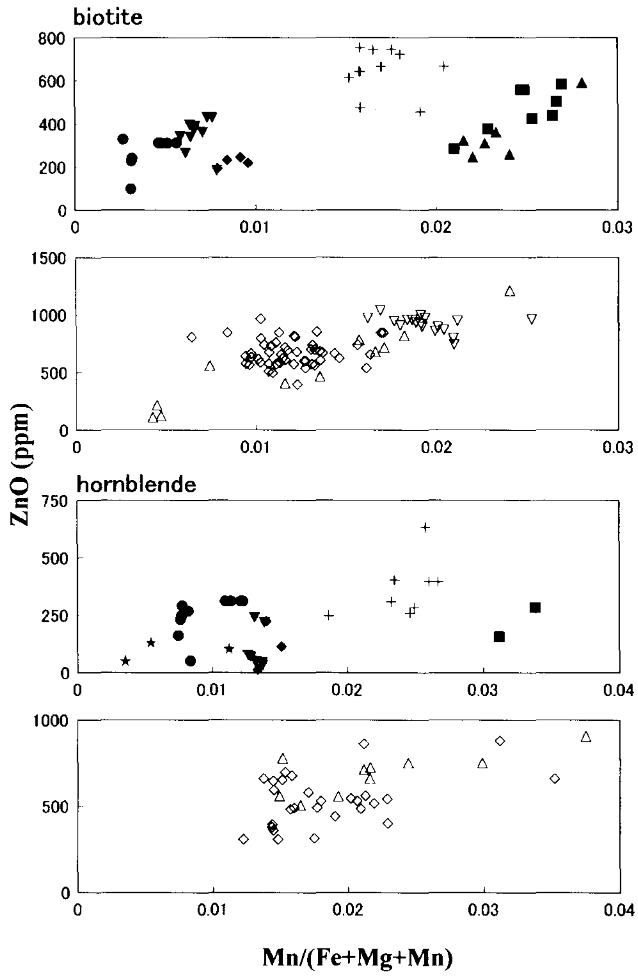

Fig. 7. Relationship between Mn/(Fe+Mg+Mn) atomic ratios and ZnO contents in mafic minerals. Symbols are the same as Fig.4.

400ppm for magnetite-series. The ZnO contents of hornblende are significantly smaller than those of biotite in both the ilmenite-series and magnetite-series granitic rocks, and they are from 304 to 902ppm, with an average value of about 600ppm in the ilmenite-series, and from 12 to 631ppm, and about 220ppm on an average in the magnetite-series.

In the ilmenite-series granitic rocks, the ZnO contents are positively correlated with the Fe/(Fe+Mg) and Mn/(Fe+Mg+Mn) ratios for biotite and hornblende (Figs. 6 and 7). The overall variation of the contents in biotite and hornblende from the magnetite-series granitic rocks, on the contrary, seems to be negatively correlated with the Fe/(Fe+Mg) ratios. However, the ZnO contents increase with increasing Fe/(Fe+Mg) ratios for each granitic body. The relationship between Mn/(Fe+Mg+Mn) ratios and ZnO contents of biotite and hornblende is ambiguous in magnetite-series. Positively correlated relationship is found between number of Mn per formula unit and the Fe/(Fe+Mg) atomic ratios for biotite and hornblende from the ilmenite-series rocks (Fig. 8). For biotite and hornblende from the magnetite-series rocks, the relationships observed between number of Mn per formula unit and the Fe/(Fe+Mg) ratios are very similar to those between ZnO contents and the Fe/(Fe+Mg) ratios. Behavior of Zn and Mn appear to be similar during the crystallization of magma. Hence, Zn contents of mafic minerals might be closely related with the Fe contents.

In the Yubara area, the amounts of mafic minerals decrease in the order of Inatsu, Yubara South, Yubara North and Unanji. Plagioclase decreases in amount relative to quartz and K-feldspar in the same order. On the contrary, K-feldspar and quartz increase in this order (Fig. 4-a). It is uncertain whether these plutonic bodies are cognate with each other or not, the ZnO contents of biotite and hornblende are higher in the apparently fractionated rocks with higher proportion of quartz and K-feldspar (Fig. 9).

The degree of fractionation must be recorded in the compositional zoning of mafic minerals. The back-scattered electron images of EPMA reveal that a compositional zoning is weakly recognized in the Kami-iwami hornblende, with an increase of the Fe/(Fe+Mg) ratios from core to rim of grains. However the variation of Zn

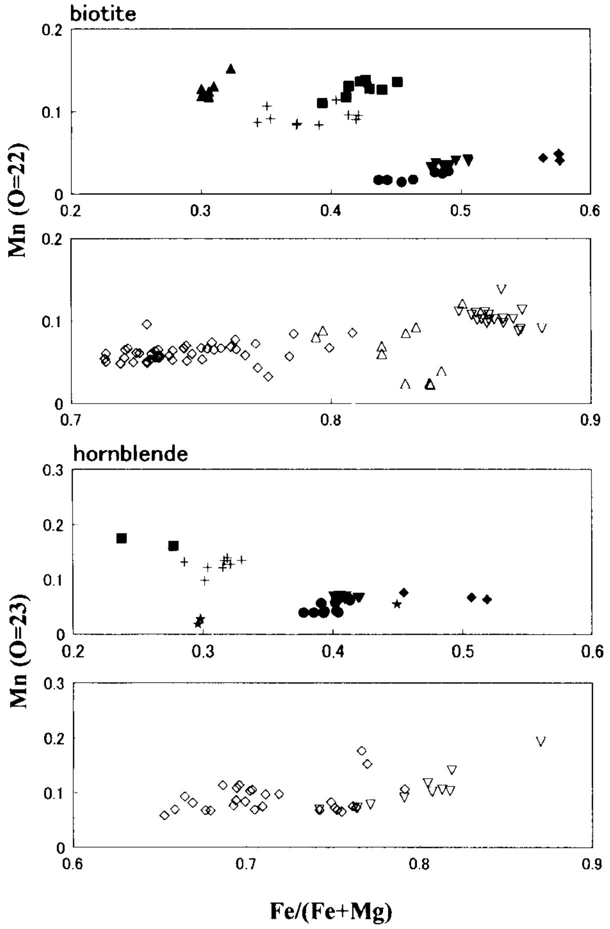

Fig. 8. Relationship between Fe/(Fe+Mg) atomic ratios and number of Mn per formula unit in mafic minerals. Symbols are the same as in Fig. 4.

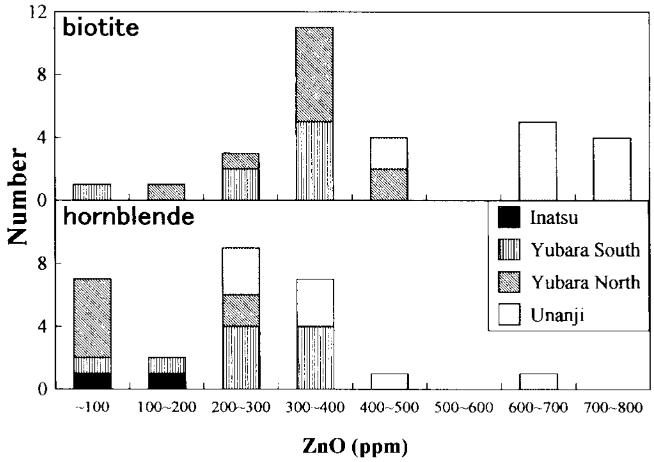

Fig. 9. Diagram showing the frequency of ZnO contents of mafic minerals from the plutonic rocks in the Yubara area. Refer to the text on the mutual relations among plutonic rocks.

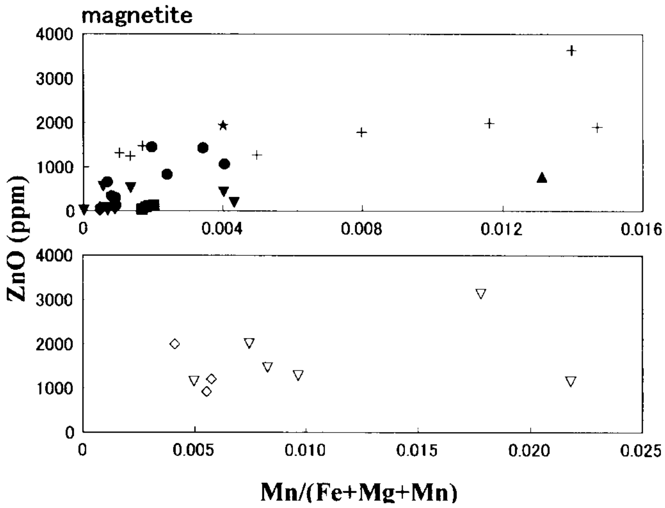

Fig. 10. Relationship between Mn/(Fe+Mg+Mn) atomic ratios and ZnO contents in magnetite. Symbols are the same as in Fig. 4.

contents are irregular independent of the Fe contents. The Fe/(Fe+Mg) ratios of the Mannari hornblende are even from core to rim of a grain, but Zn contents slightly decrease toward rim. Compositional zoning is hardly detected for biotite from Kami-iwami and Mannari by back-scattered electron images.

2. magnetite

The Zn contents in magnetite from magnetite-series granitic rocks are usually low (smaller than about 120ppm). Exceptionally, magnetite from the Unanji and some Yubara South samples has high Zn contents, ranging from 1230 to 3620ppm. The Mn/(Fe+Mg+Mn) ratios of Zn-poor magnetite from the Kami-iwami, Yokota, Sangunsan and Yubara North masses range from 0 to 0.004, their average being 0.001. On the other hand, those of Zn-rich magnetite from Yubara South and Unanji are somewhat larger, ranging from 0.002 to 0.006, with an average value of 0.003, and from 0.001 to 0.012 with 0.007 average value, respectively. Figure 10 shows that the ZnO contents (ppm) of magnetite increase with increasing the Mn/(Fe+Mg+Mn) atomic ratios. The ZnO contents of magnetite occurring in trace amounts in the ilmenite-series rocks, range from 768 to 3118ppm, with an average value of 1575ppm. The contents are somewhat larger than those from the magnetite-series rocks.

3. Zn/Fe ratios

The Zn contents of mafic minerals appear to be closely related with the Fe contents, as described previously. In particular, the correlation relation is more distinctly observed in the ilmenite-series rocks. It is therefore significant to compare the Zn/Fe ratios between ilmenite-series and magnetite-series granitic rocks in order to know the behavior of Zn during the crystallization of magma. Figure 11 is a histogram of the Zn/Fe ratios of hornblende, biotite and magnetite. The Zn/Fe ratios of hornblende are about 2.10×10^{-3} for magnetite-series, and about 2.28×10^{-3} for ilmenite-series. The ratios of biotite are about 1.35×10^{-3} for magnetite-series, and about 1.90×10^{-3} for ilmenite-series. The Zn/Fe ratios seem to be slightly larger in the ilmenite-series granitic rocks for both biotite and hornblende. The Zn/Fe ratios of magnetite from magnetite-series are about 0.75×10^{-3} in average, distinctly smaller than those of biotite and hornblende. Larger Zn/Fe ratios of magnetite are mainly obtained from the Unanji sample, which has high Mn contents as mentioned above.

VI. Discussion

Microscopic observations revealed that no sphalerite is found in studied polished sections from magnetite-series granitic rocks (Fig. 3). Hence, there are no microscopic evidences that magmas that brought about magnetite-series granitic rocks were saturated in ZnS during the crystallization.

As mentioned previously, ilmenite-series granitic rocks contain mafic minerals with Fe/(Fe+Mg) and Zn/Fe ratios larger than those in magnetite-series. Along with the progress in crystallization, ilmenite-series granitic magmas probably become poorer in Zn, compared with the magnetite-series granitic magmas. However no criteria could be deduced from these chemical data that Zn contents or Zn/Fe ratios increase in the residual magma accompanied with the progress in crystallization.

It is needed to assess the effect of magnetite crystallization on the enrichment or depletion of Zn in the magma, because an abundant occurrence of magnetite, probably in the early crystallization stage, is a distinct mineralogical feature discriminating the magnetite-series from the ilmenite-series granitic rocks. The Zn/Fe ratios of magnetite are generally smaller than 2.5×10^{-3}, with an average of about 0.75×10^{-3}. In addition the values are smaller than those of biotite and hornblende (Fig. 11). The modal abundance of magnetite in the magnetite-series granitic rocks is only a few percents of the sum of biotite and hornblende, thus it is hardly assumed that the crystallization of magnetite depletes the Zn contents in the magma. Of course, the effect of magnetite is ignored for its trace amounts in the ilmenite-series magma. These discussions suggest that the magnetite-series granitic magma becomes richer in Zn contents relative to those of ilmenite-series magma in the later stage of the crystallization.

Urabe (1985,1987) experimentally investigated the partitioning of Zn between melts and fluids, and indicated that Zn is strongly partitioned in hydrothermal fluid. The experimental result indicates that the fluids derived from fractionated magnetite-series granitic magma appear to be enriched in Zn relative to those from ilmenite-series one, thus magnetite-series granitic magma is considered to be favorable for the formation of Zn-rich deposits.

VII. Conclusion

1. Sphalerite generally does not occur even in the magnetite-series granitic rocks associated with Zn-Pb-rich deposits in the Inner Zone of Southwest Japan, indicating that the magnetite-series granitic magma was undersaturated in ZnS during the crystallization process.

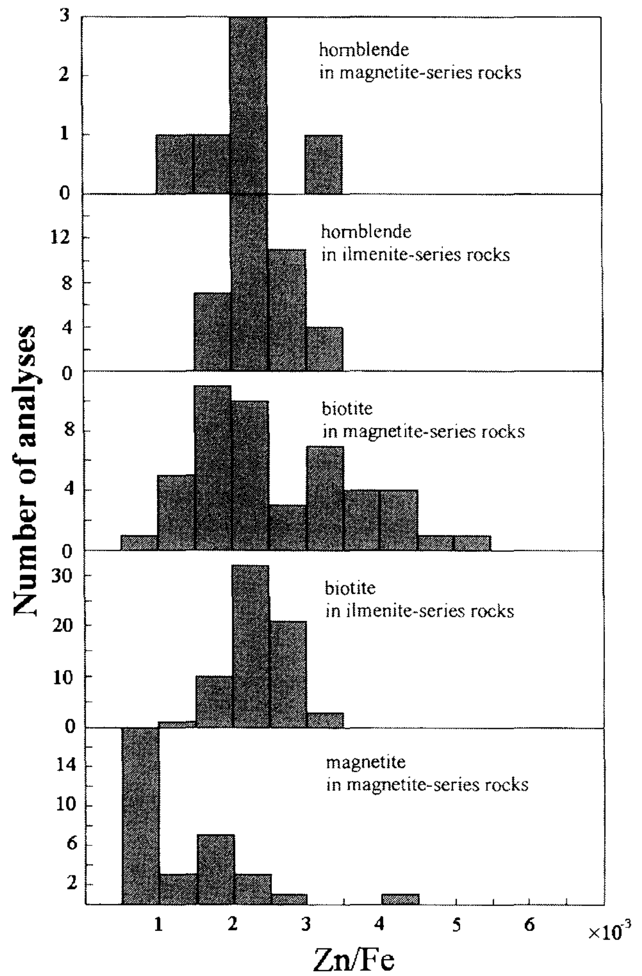

Fig. 11. The Zn/Fe atomic ratios of mafic minerals in granitic rocks.

2. The mafic minerals such as biotite and hornblende in the ilmenite-series granitic rocks have markedly larger Fe/(Fe+Mg) ratios and slightly larger Zn/Fe ratios than those in the magnetite-series. Therefore Zn is considered to be much more efficiently removed by the crystallization of mafic minerals in the ilmenite-series granitic magma. Fractionated ilmenite-series granitic magma is probably poorer in Zn compared with the correspondingly fractionated magnetite-series magma.
3. It is needed to assess the role of magnetite crystallized from the magnetite-series magma on the removal of Zn. The Zn/Fe ratios of magnetite were found to be commonly smaller than those of biotite and hornblende, and the modal abundance is usually only several percents of those of biotite and hornblende. Therefore it is concluded that crystallization of magnetite does not play an important role for the removal of Zn from the magnetite-series magma.

Acknowledgement: Sincere thanks are due to Associate Prof. Toshihiko Matsuda, Department of Earth Sciences, Okayama University, for his kind instruction in the use of the X-ray diffractometer. Technical assistance provided by Messrs. Toshiaki Saito and Natsuya Ando of the Department of Earth Sciences is greatly acknowledged. One of the authors (H.T.) would like to thank the colleagues of the Laboratory for Mineral Deposit Research, Department of Earth Sciences, for their kind encouragement.

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