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Fe-Ti OXIDE MINERALS IN THE MATSUMAE PLUTONIC ROCKS, SOUTHWESTERN HOKKAIDO, JAPAN

by

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(with 6 text-figures and 3 tables)

Abstract

The Matsumae plutonic rocks, consisting of ultramafic cumulates to adamellite, are divided into three types in terms of the assemblage 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. Chemistry of Fe-Ti oxide minerals suggests that origin of these types is ascribed to the difference in oxidation state of magma, i.e. Type I is the highest in f_{O_2} while Type III is the lowest. However, these differences in f_{O_2} do not strictly correlate with the variation of assumed f_{H_2O}.

Some rocks of Type II may represent an original redox state of the Matsumae plutonic magma. The Type I rocks were probably oxidized mainly in the magma chamber, whereas the Type III rocks may be reduced. Since some of the hornblendite show mixed facies of Types I and II, incorporation of less oxidized magma into the oxidized magma chamber may have occurred in early stages of crystallization. Redox state of the Matsumae plutonic magma is considered to be affected by such magma incorporation.

Introduction

Oxidation state of igneous rocks have been studied by many authors. In particular, Ishihara (1977) divided granitic rocks into magnetite- and ilmenite-series in terms of the presence or absence of magnetite. Since these two series indicate the difference in oxidation state of magma, this division is important to investigate petrogenesis and metallogenesis of granitic rocks (Ishihara, 1977).

Concerning the Cretaceous plutonic rocks in Northeast Japan, those in the Kitakami belt are characterized by the predominance of the magnetite-series, whereas those in the Abukuma belt by the ilmenite-series. Cretaceous plutonic rocks in southwestern Hokkaido, though the data are not sufficient, appear to consist mainly of ilmenite-series rocks likely in the Abukuma belt (Ishihara, 1979). The Matsumae plutonic rocks in southwestern Hokkaido, however, consist mainly of the magnetite-series rocks in contrast with other plutonic rocks in the same region (Tsuchiya, 1982). In addition, the Matsumae plutonic rocks indicate various oxidation states from typical magnetite-series rocks to ilmenite-series ones, suggesting complicated evolution of redox state of the magma.

In this paper, the author reports the mode of occurrence and chemistry of Fe-Ti oxide minerals, and discusses the oxidation state of the Matsumae plutonic magma.

Outline of petrography and petrochemistry

The Matsumae plutonic rocks consist of nine rock types which range from

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ultramafic cumulates to adamellite; clinopyroxenite, hornblendite I, hornblendite II (these three rock types are considered to be cumulate judging from the texture), granodiorite, aplite I, monzogabbro, quartz monzodiorite, adamellite, and aplite II. The former five rock types are characterized by crystallization of amphibole prior to plagioclase, and are named 'Am series'. On the other hand, the crystallization sequence in the latter four rock types is reverse and is named 'P1 series' (Tsuchiya, in press). The two series are considered to indicate the difference in f_{H_2O} in magma; the Am series seems to be higher in f_{H_2O} than the P1 series.

These rocks are characterized by high alkali, especially K_2O content, but the Am series is slightly less alkalic than the P1 series owing to amphibole fractionation. Detailed petrography and petrochemistry are described in Tsuchiya (1982, in press).

Description of Fe-Ti oxide minerals

The Matsumae plutonic rocks are divided into Type I to III in terms of the characteristics of Fe-Ti oxide minerals (Table 1). Type I is characterized by the predominance of magnetite and the presence of independent ilmenite with hematite lamellae (hemo-ilmenite of Buddington et al., 1963). Composite grains of ilmenite and magnetite (ilmeno-magnetite, ibid.) are usually observed. Coexisting sulfides are pyrite and subordinate chalcopyrite. Sphene and epidote commonly occur as accessories. Oxidation products of hematite after magnetite, and hematite and rutile after ilmenite are occasionally observed. This type is assigned to the typical magnetite-series of Ishihara (1977). Most of the P1 series rocks and some of the Am series correspond to Type I.

Table 1 Assemblage of opaque minerals in each rock type

		modal %	Mt	cIl	iIl	lHm	Pl	Cr	Py	Po	Cp
Type I	hornblendite I	1.6— 5.8	○	○	+	+	(+)	(+)	○	−	+
	granodiorite	— 0.9	○	+	+	+	−	(+)	−	−	+
	monzogabbro	— 2.2	○	○	+	−	−	−	−	−	+
	qz. monzodiorite	0.8— 3.3	○	+	(+)	(+)	−	−	(+)	(+)	+
	adamellite	0.3— 1.4	○	+	(+)	?	−	−	−	−	+
Type II	clinopyroxenite	1.5—14.2	○	○	−	−	+	(+)	(+)	+	+
	hornblendite I	— 2.0	+	○	+	−	(+)	(+)	○	(+)	+
	hornblendite II	0.4— 5.6	○	○	+	−	(+)	−	○	(+)	+
	granodiorite	0.1— 0.6	+	+	+	−	−	(+)	○	−	+
	monzogabbro	2.9— 4.0	○	○	+	−	−	−	−	−	+
Type III	granodiorite	— 2.2	−	−	+	−	−	−	(+)	○	+

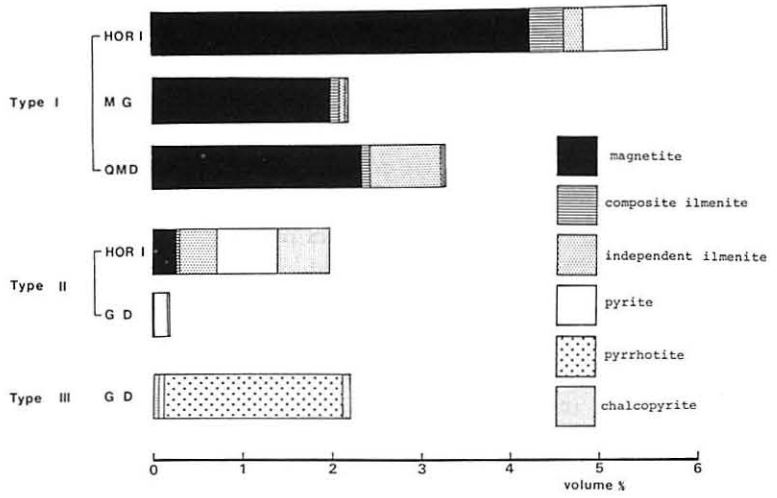
○: remarkable, +: common, (+): sometimes observed, −: nil
mt: magnetite, cIl: composite ilmenite, iIl: independent ilmenite, lHm: lamellar hematite,
Pl: pleonaste, Cr: chromite, Py: pyrite, Po: pyrrhotite,
Cp: chalcopyrite.

Type II is characterized by the absence of hemo-ilmenite, and is generally poorer in magnetite than Type I. Coexisting sulfides and accessories are similar to Type I, but pyrrhotite is contained in some specimens. This type is classified into the magnetite-

series of Ishihara, though it is considered to be lower in f_{O_2} than Type I as described below. Many rocks of the Am series and some of the monzogabbro of the P1 series are classified into this type.

Type III is characterized by the absence of magnetite. Coexisting sulfides are pyrrhotite with minor amounts of chalcopyrite and trace of pyrite. Sphene is observed as a common accessory. This type has the nature of typical ilmenite-series of Ishihara except for high modal abundance of opaque minerals and sphene. Some rocks of the melanocratic granodiorite correspond to this type, but their occurrence is limited to a narrow area.

Modal analyses of opaque minerals in the representative specimens were carried out after the method of Itaya and Banno (1980), and the results are shown in Text-fig. 1. Since the aplite I and II contain only trace amounts of opaque minerals, these aplites are not discussed here.



Text-fig. 1 Modal compositions of opaque minerals in representative specimens.

The ilmenite occurs as independent or composite grains intergrown with magnetite. For description of composite type ilmenite, the terms of trellis, sandwich, and granular of Buddington and Lindsley (1964) and Haggerty (1976) are used. Trellis ilmenite, however, occasionally grades into sandwich type, and the division between them is rather obscure in the Matsumae plutonic rocks. In such cases, ilmenite lamellae of platy occurrence are treated as the trellis type.

Clinopyroxenite

The clinopyroxenite is classified into Type II. This rock contains 1.5-14.2 vol.% of of opaque minerals, and magnetite predominates in them. Magnetite (100-1000μm) usually occurs as intercumulus minerals, and it is sometimes observed as granular

grains (10-100μm) in clinopyroxene and amphibole. The magnetite occasionally has lamellae of ilmenite and pleonaste (Text-fig. 2-1 and 2-2). The ilmenite occurs as granular, sandwich, and trellis intergrowth, but independent ilmenite is not observed. The granular ilmenite is observed as irregular grains (up to 100μm) and the sandwich ilmenite as thin plates (20-50μm) in the magnetite grains. The trellis ilmenite generally occurs as thin plates (1-5μm), and some of them grade into the sandwich type.

The pleonaste is occasionally observed as small irregular blebs (0.5-5μm, 20μm in maximum), and is sometimes concentrated around ilmenite lamellae and margins of the magnetite grains. Anhedral chalcopyrite and granular pyrrhotite are usually included.

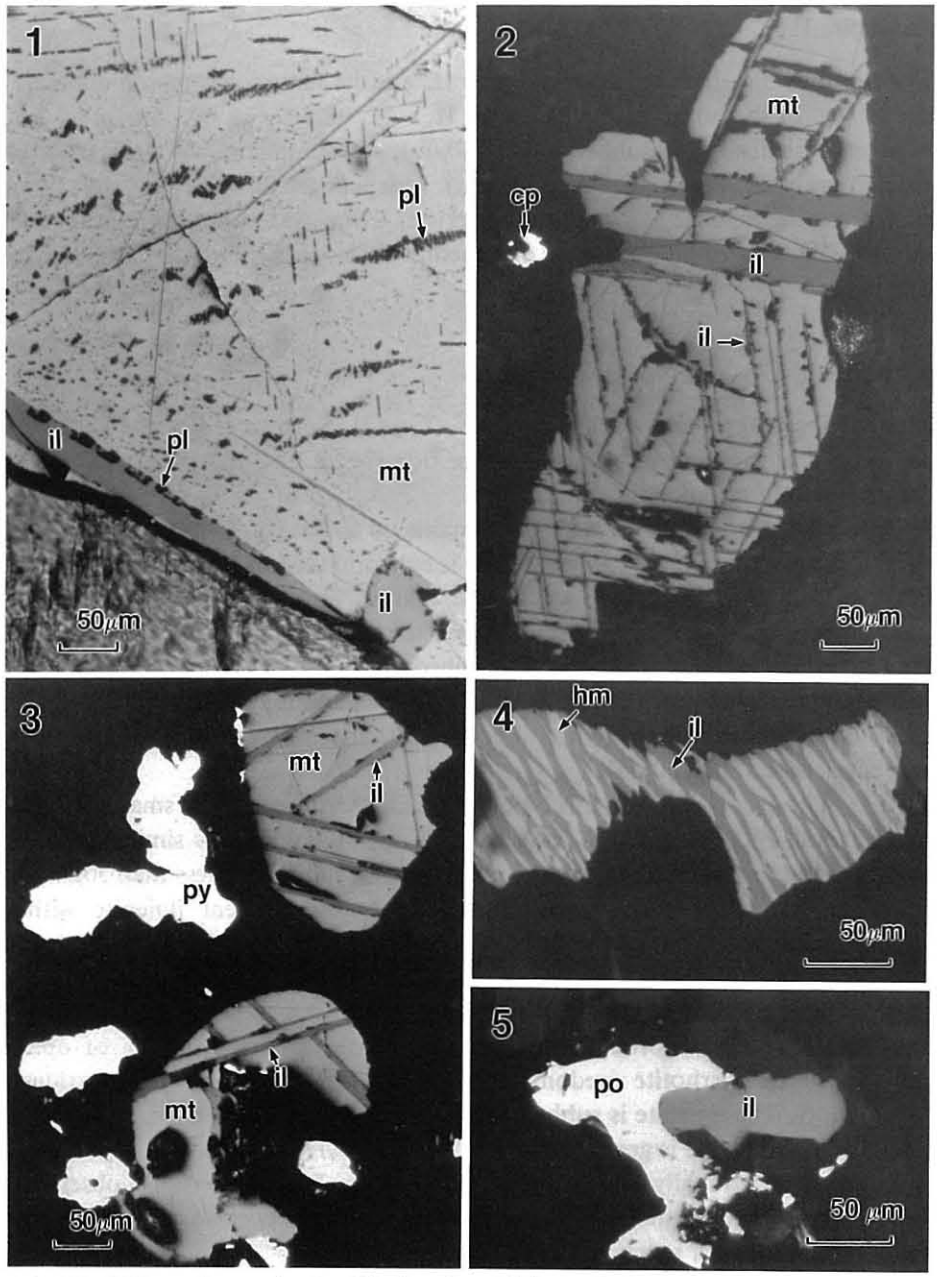
Some specimens of the clinopyroxenite carry Cr-diopside (Tsuchiya, in press), which rarely contain small granular chromite (less than 50μm).

Hornblendite I

The hornblendite I belongs mainly to Type II, and some of them to Type I. In rare cases (No. 52706), the lithologies of Types I and II coexist and are mixed irregularly in a single specimen.

Type I; The hornblendite I of this type contains 1.6-5.8 vol.% of opaque minerals in which magnetite predominates. Magnetite is euhedral to subhedral (50-500μm), and generally has ilmenite intergrowths and sometimes pleonaste lamellae. The magnetite rarely includes granular chromite cores (50-200μm). The ilmenite intergrowth is observed as granular, sandwich, and trellis lamellae. The granular ilmenite shows irregular shape (up to 100μm) and rarely has hematite lamellae. The sandwich ilmenite (10-30μm) shows irregular plates and sometimes grades into granular ilmenite. The trellis ilmenite occurs as thin plates (less than 5μm). Independent hemo-ilmenite (20-500μm) is subhedral to euhedral and has thin irregular plates (less than 2μm) of hematite. It is occasionally intergrown with sphene. In places, these ilmenites are oxidized into hematite and rutile. Coexisting sulfides are pyrite and subordinate chalcopyrite. They are generally anhedral (less than 200μm) and occupy the interstices. The pyrite, however, is rarely observed as granular grains (10-100μm) included in amphibole or magnetite.

Type II; The hornblendite I of this type contains smaller amounts of opaque minerals (less than 2.0 vol.%) than that of Type I. Magnetite is euhedral to subhedral (20-200μm) and occasionally has trellis (less than 10μm) and sandwich (10-20μm) ilmenite with minor amounts of pleonaste. Minor amounts of granular ilmenite are also observed in the magnetite. The magnetite rarely includes granular chromite core. Independent ilmenite is subhedral to anhedral (20-100μm) and has no hematite lamellae. Some of the subhedral ilmenites aggregate with chlorite and/or sphene. Sulfide minerals are anhedral to granular pyrite, subordinate amounts of anhedral chalcopyrite, and rare granular pyrrhotite.



Text-fig. 2 Photomicrographs of polished sections (oil immersion).
1: Magnetite with granular ilmenite and minute blebs of pleonaste (clinopyroxenite, Type II).
2: Subhedral magnetite with sandwich and trellis ilmenite (hornblendite I, Type II).
3: Subhedral magnetite with trellis ilmenite and anhedral pyrite (hornblendite I, Type I).
4: Anhedral ilmenite with hematite lamellae (monzogabbro, Type I).
5: Subhedral ilmenite intergrown with anhedral pyrrhotite (granodiorite, Type III).
mt: magnetite, il: ilmenite, pl: pleonaste, cp: chalcopyrite, py: pyrite, hm: hematite, po: pyrrhotite.

Hornblendite II

The hornblendite II is regarded as Type II, and its nature of opaque minerals is similar to that of the hornblendite I of Type II.

Magnetite is euhedral to subhedral (20-100μm), and sometimes has trellis (less than 10μm) and granular ilmenite (less than 20μm). Independent ilmenite (less than 50μm) without hematite lamellae is sometimes observed. Coexisting sulfides are granular to anhedral pyrite, subordinate amounts of anhedral chalcopyrite, and rare granular pyrrhotite.

Granodiorite

Most of the granodiorites are regarded as Type II. Some rocks of the leucocratic granodiorite are classified into Type I. Rare occurrence of Type III granodiorite is locally observed in melanocratic part.

Type I; The granodiorite of this type contains up to 0.9 vol.% of opaque minerals in which magnetite predominates. Magnetite is euhedral to subhedral (10-100μm), and usually oxidized into hematite around the margin and along the cleavage. The magnetite occasionally has granular ilmenite (less than 20μm). Independent hemo-ilmenite is rarely observed as subhedral to anhedral grains (20-50μm). These ilmenites are generally oxidized into hematite and rutile. Coexisting sulfides are accessory amounts of pyrite and chalcopyrite. Very rarely, the magnetite contains granular chromite cores (less than 50μm).

Type II; The granodiorite of this type contains a characteristically small amount of opaque minerals (0.1-0.6 vol.%). The nature of opaque minerals is similar to that of the hornblendite I of Type II. Magnetite is euhedral to subhedral (less than 50μm) and generally has granular ilmenite (less than 10μm). Independent ilmenite without hematite lamellae is subhedral to euhedral (less than 20μm). Coexisting sulfides are anhedral to granular pyrite and subordinate chalcopyrite. Very rarely, granular chromite cores are found in the magnetite.

Type III; The granodiorite of this type carries less than 2.2 vol.% of opaque minerals in which pyrrhotite predominates. Only ilmenite is observed as oxides in minor amounts. The ilmenite is subhedral to euhedral with no hematite lamellae (less than 50μm). The ilmenite is generally intergrown with pyrrhotite and/or sphene (Text-fig. 2-5) and is occasionally included in the amphibole. Pyrrhotite is also observed as anhedral to subhedral crystal. Subhedral to anhedral chalcopyrite and trace amounts of pyrite associated with pyrrhotite are also present.

Monzogabbro

Coarse grained rocks of the monzogabbro are regarded as Type I, whereas fine grained ones are Type II.

Type I; The monzogabbro of this type contains up to 2.2 vol.% of opaque

minerals, and magnetite predominates in them. Magnetite is euhedral to subhedral (10-200μm), and contains granular ilmenite (less than 50μm) and minor amounts of sandwich (less than 30μm) and trellis (less than 5μm) ilmenite. Independent ilmenite is usually anhedral (20-200μm), and is included in biotite or occupies interstices. The independent ilmenite characteristically contains relatively thick irregular plate of hematite lamellae (5-20μm, Text-fig. 2-4). Accessory amounts of chalcopyrite are also observed. Magnetite showing vermicular intergrowth with orthopyroxene (1-5μm) sometimes surrounds olivine. Similar occurrence is described in some gabbroic rocks and is considered to be an oxidation product of olivine (e.g. Ambler and Ashley, 1977).

Type II; The monzogabbro of this type contains 2.9-4.0 vol.% of opaque minerals, whose characteristics are similar to those of the monzogabbro of Type I except for the absence of hemo-ilmenite. Isolated ilmenite (20-50μm) has no hematite lamellae and is anhedral to subhedral grains which are generally included in biotite.

Quartz monzodiorite

The quartz monzodiorite carries 0.8 to 3.3 vol.% of opaque minerals, and magnetite predominates in them. Some specimens of this rock contain independent hemo-ilmenite. All of the quartz monzodiorite, however, are classified into Type I, because there is no significant difference in nature of Fe-Ti oxide minerals between hemo-ilmenite-bearing rocks and -free ones.

Magnetite is euhedral to subhedral (10-300μm), and is occasionally oxidized into hematite around the margin and along the cleavage. It has minor amounts of granular (less than 100μm) and trace amounts of trellis (less than 5μm) ilmenite. Independent ilmenite is subhedral to euhedral (50-100μm) and has hematite lamellae (less than 5μm). These ilmenites are occasionally oxidized into hematite and rutile. Minor amounts of chalcopyrite and pyrite are contained, and pyrrhotite is rarely observed.

Adamellite

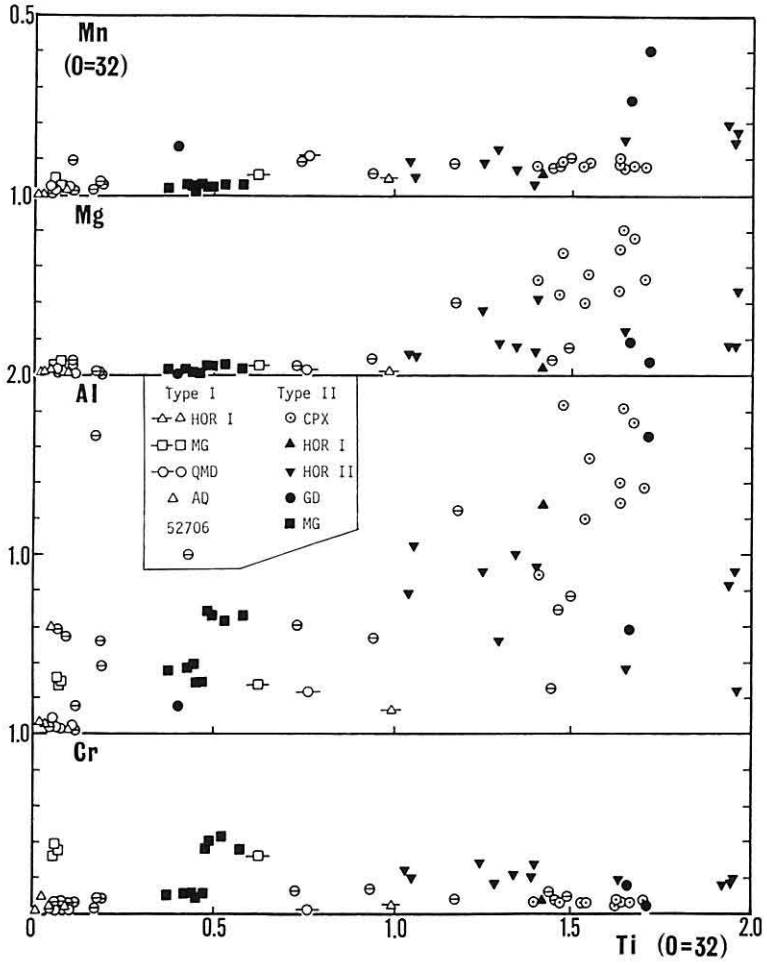
Opaque minerals in the adamellite (0.3-1.4 vol.%) are mostly oxidized, and their original nature is not well known. However, this rock can be classified into Type I because the nature of Fe-Ti oxide minerals are similar to those of the quartz monzodiorite.

Magnetite is euhedral to subhedral (50-500μm), and is generally oxidized into hematite around the margin and along the cleavage. The magnetite rarely contains granular ilmenite (less than 100μm). Independent ilmenite is rarely observed as subhedral to euhedral crystal (50-200μm). These ilmenites are generally oxidized into hematite and rutile.

Chemistry of Fe-Ti oxide minerals

Chemical analyses were carried out using the electron microprobe JEOL JXA-50A (Dept. Metallurgical Engineering, Hokkaido Univ.). The correction was made by the method of Bence and Albee (1968). Bulk composition of Fe-Ti oxide minerals with lamellae is analysed by traversing with a beam of 5 to 20μm. Bulk compositions of some magnetite in the Type I rocks cannot be obtained by such method, because of the scatter of ilmenite lamellae. In this case, the bulk composition is calculated using chemical composition of host magnetite and lamellar ilmenite and their modal abundance.

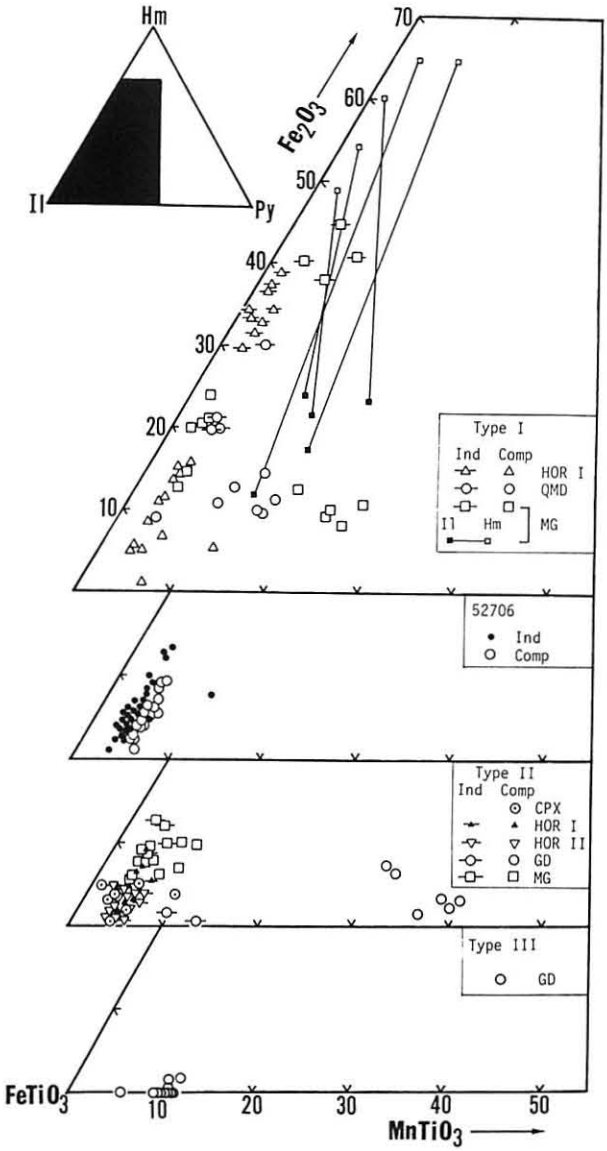
Table 2 shows representative analyses of magnetite. Fe_2O_3 content of magnetite is



Text-fig. 3 Mn, Mg, Al, and Cr vs. Ti relationships of the magnetite.
Symbols with horizontal bar represent calculated composition of ilmenomagnetite. Abbreviations are the same as in Table 2.

calculated assuming stoichiometry, and ulvöspinel mol% of Fe-Ti two component system (X'usp in Table 1) is estimated by the method of Stormer (1983).

Text-fig. 3 shows Mn, Mg, Al, and Cr vs. Ti relationships of the magnetite. The magnetite in the Type I rocks is poor in these elements, and usually has the composition of almost pure magnetite. Calculated composition of the magnetite host plus ilmenite lamellae is rather richer in Ti. In the Type II rocks, the magnetite is generally rich in Ti except for the monzogabbro. Al and Mg show weak positive correlation with Ti. Mn and Cr are generally low and show no correlation with Ti, but the magnetite in the



Text-fig. 4 $FeTiO_3$-$MnTiO_3$-Fe_2O_3 diagram of the ilmenite.
Il: ilmenite, Py: pyrophanite, Hm: hematite, Ind: independent grain, Comp: composite grain. Other abbreviations are the same as in Table 2.

Table 2 Representative analyses of the magnetite

	1 HOR I 52703	2 MG 52210	3 QMD 52901	4 AD 52109	5 CPX 52201	6 HOR I 52706B	7 HOR II 52513	8 GD 52810	9 MG 52508	10 HOR I 52703	11 CPX 52201
SiO_2	0.06	n.a.	n.a.	n.a.	n.a.	0.10	2.64	1.16	n.a.	0.09	n.a.
TiO_2	4.21	2.72	3.22	0.09	6.93	6.14	8.50	7.19	2.50	0.13	0.43
Al_2O_3	0.36	0.78	0.63	0.08	4.37	3.50	0.64	1.56	1.81	8.11	61.91
Cr_2O_3	0.24	1.37	0.07	0.24	0.24	0.28	0.80	0.64	1.48	54.03	0.40
FeO*	87.24	88.87	87.74	92.12	81.55	82.19	78.72	80.85	87.52	34.61	18.41
MnO	0.18	0.25	0.43	0.15	0.37	0.23	0.67	1.01	0.11	0.56	0.13
NiO	0.02	n.a.	n.a.	n.a.	n.a.	0.05	0.05	0.01	n.a.	tr.	n.a.
MgO	0.03	0.12	0.04	tr.	1.26	0.09	1.04	0.39	0.09	2.13	15.61
CaO	0.01	n.a.	n.a.	n.a.	n.a.	0.06	0.11	0.17	n.a.	0.01	n.a.
Sum	92.35	94.11	92.13	92.68	94.72	92.64	93.17	92.98	93.51	99.67	96.89
Fe_2O_3	58.85	61.68	60.74	68.12	50.76	50.99	43.72	48.67	60.25	5.40	1.32
FeO	34.29	33.37	33.09	30.83	35.87	36.30	39.38	37.06	33.31	29.75	17.22
Total	98.25	100.29	98.22	99.51	99.80	97.74	97.55	97.86	99.55	100.21	97.02
					Cations per 32 oxygens						
Si	0.019	—	—	—	—	0.031	0.807	0.356	—	0.025	—
Ti	0.987	0.623	0.755	0.021	1.545	1.416	1.954	1.660	0.574	0.028	0.069
Al	0.132	0.280	0.231	0.029	1.527	1.265	0.231	0.565	0.651	2.697	15.582
Cr	0.059	0.330	0.017	0.059	0.056	0.068	0.193	0.155	0.357	12.052	0.068
Fe^{3+}	13.798	14.143	14.242	15.870	11.326	11.772	10.055	11.247	13.843	1.146	0.212
Fe^{2+}	8.935	8.504	8.622	7.982	8.895	9.314	10.065	9.517	8.505	7.020	3.076
Mn	0.048	0.065	0.113	0.039	0.093	0.060	0.173	0.263	0.028	0.134	0.024
Ni	0.005	—	—	—	—	0.012	0.012	0.002	—	—	—
Mg	0.014	0.055	0.019	—	0.557	0.041	0.474	0.179	0.041	0.896	4.970
Ca	0.003	—	—	—	—	0.020	0.036	0.056	—	0.003	—
Total	24.000	24.000	23.999	24.000	23.999	23.999	24.000	24.000	23.999	24.001	24.001
X'usp	12.58	8.31	9.60	0.26	22.47	20.89	27.48	22.98	8.11	—	—

n.a.: not analysed, tr.: not detectd.
CPX: clinopyroxenite, HOR I: hornblendite I, HOR II: hornblendite II, GD: granodiorite, MG: monzogabbro,
QMD: quartz monzodiorite, AD: adamellite, X'usp: ulvöspinel molecule calculated by the method of Stormer (1983). 1-4: Type I, 5-9: Type II, 10: chromite core in the magnetite, 11: pleonaste lamellae in the magnetite, 1-3: calculated bulk compositions of ilmeno-magnetite, 5-9: bulk composition of magnetite + pleonaste lamellae obtained by a broad beam, 4, 10, and 11: point analysis.

Table 3 Representative analyses of the ilmenite

	1 HOR I 52703	2 HOR I 52703	3 MG 52210	4 MG 52210	5 QMD 52901	6 QMD 52901	7 CPX 818R	8 HIR I 52706B	9 HOR II 52513	10 GD 52810	11 GD 52810	12 MG 52508	13 GD 80504
SiO_2	0.06	0.02	n.a.	n.a.	n.a.	n.a.	0.23	0.12	0.09	0.44	1.09	n.a.	0.08
TiO_2	31.28	44.07	28.25	48.05	40.83	46.59	53.43	50.25	51.24	50.84	49.58	49.97	52.68
Al_2O_3	0.03	0.01	0.05	0.07	0.07	0.02	0.01	0.02	0.05	0.10	0.02	0.05	0.02
Cr_2O_3	0.05	0.11	0.51	0.16	0.01	tr.	0.21	0.17	tr.	tr.	0.14	0.13	tr.
FeO*	62.95	51.56	63.87	39.51	51.88	45.58	36.33	46.43	45.94	42.02	28.87	47.24	41.02
MnO	0.67	1.76	2.19	11.07	2.09	5.90	3.53	1.59	2.35	4.69	17.79	1.81	5.67
NiO	tr.	tr.	n.a.	n.a.	n.a.	n.a.	0.06	tr.	tr.	tr.	0.08	n.a.	0.04
MgO	0.09	0.07	0.07	0.08	0.37	0.05	6.33	0.11	0.08	0.10	0.04	0.29	0.07
CaO	0.03	0.01	n.a.	n.a.	n.a.	n.a.	0.01	0.09	0.19	0.48	0.77	n.a.	0.11
Sum	95.16	97.61	94.94	98.94	95.25	98.14	100.14	98.78	99.94	98.67	98.38	99.49	99.69
Fe_2O_3	39.60	15.37	45.35	8.51	19.94	10.84	3.27	3.36	2.80	1.47	2.37	5.18	—
FeO	27.32	37.73	23.06	31.85	33.94	35.83	33.39	43.41	43.42	40.70	26.73	42.58	41.02
Total	99.13	99.15	99.48	99.79	97.25	99.23	100.47	99.12	100.22	98.82	98.61	100.01	99.69
Cations per 3 oxygens													
Si	0.002	0.001	—	—	—	—	0.006	0.003	0.002	0.011	0.028	—	0.002
Ti	0.611	0.850	0.551	0.916	0.803	0.895	0.963	0.963	0.970	0.973	0.948	0.949	1.000
Al	0.001	—	0.002	0.002	0.002	0.001	—	0.001	0.001	0.003	0.001	0.001	0.001
Cr	0.001	0.002	0.010	0.003	—	—	0.004	0.003	—	—	0.003	0.003	—
Fe^{3+}	0.774	0.297	0.886	0.162	0.392	0.208	0.059	0.064	0.053	0.028	0.045	0.098	—
Fe^{2+}	0.593	0.809	0.500	0.675	0.742	0.766	0.669	0.925	0.914	0.866	0.568	0.899	0.866
Mn	0.015	0.038	0.048	0.238	0.046	0.128	0.072	0.034	0.050	0.101	0.383	0.039	0.121
Ni	—	—	—	—	—	—	0.001	—	—	—	0.002	—	0.001
Mg	0.003	0.003	0.003	0.003	0.014	0.002	0.226	0.004	0.003	0.004	0.002	0.011	0.003
Ca	0.001	—	—	—	—	—	—	0.002	0.005	0.013	0.021	—	0.003
Total	2.001	2.000	2.000	1.999	1.999	2.000	2.000	1.999	1.998	1.999	2.001	2.000	1.997
X'il	60.87	84.81	54.23	90.66	79.75	88.84	96.45	96.72	97.26	98.50	97.02	94.96	100.00

n.a.: not analysed, tr.: not detected.

1-6: Type I, 7-12: Type II, 13: Type III, X'il: ilmenite molecule calculated by the method of Stormer (1983).

1, 3, and 5: bulk composition of hemo-ilmenite obtained by a broad beam, 9, 10, and 13: independent ilmenite, others: composite ilmenite.

granodiorite is relatively rich in Mn. The magnetite in the clinopyroxenite is generally richer in Mg and Al and poorer in Cr than the other Type II rocks.

Some magnetites in the hornblendite II have a considerably high SiO_2 content up to 2.8 wt% (analyses by broad beam). Such high Si content of magnetite is rarely reported from some volcanic rocks (Imaoka et al., 1982).

Table 3 shows representative analyses of the ilmenite. Recalculation procedure is the same as in the case of the magnetite. Most of the ilmenites are approximated by a $FeTiO_3$-$MnTiO_3$-Fe_2O_3 three component system, though some ilmenite in the clinopyroxenite are rather high in MgO (up to 6 wt% MgO). Text-fig. 4 is $FeTiO_3$-$MnTiO_3$-Fe_2O_3 diagram. The ilmenite of each type is clearly distinguished in this diagram.

Independent hemo-ilmenite in the Type I rocks is characterized by considerably high Fe_2O_3 component. Hematite lamellae of hemo-ilmenite are slightly poorer in $MnTiO_3$ than the host ilmenite. Composite ilmenite in the Type I rocks shows a wide compositional range, and is generally richer in $MnTiO_3$ and Fe_2O_3 than the rocks of Types II and III.

The ilmenite in the Type II rocks is rather poorer in Fe_2O_3 than in the Type I rocks. The composite ilmenite in the granodiorite is extremely rich in $MnTiO_3$. In the Type II rocks, there is no significant difference in Fe_2O_3 between independent and composite ilmenites. In the specimen No. 52706, which consists of mixed facies of Types I and II, both composite and independent ilmenites show a wide variation in Fe_2O_3 within a single specimen.

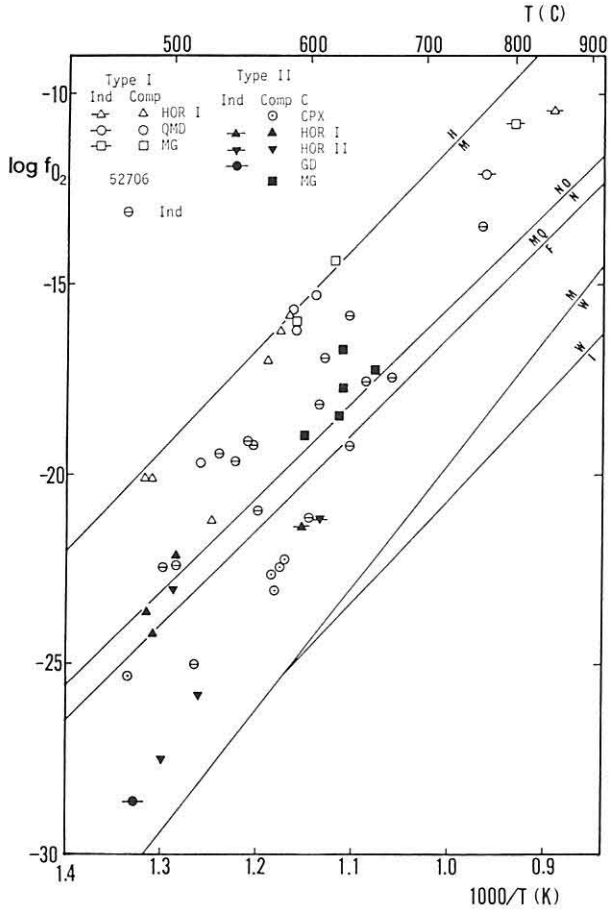
The ilmenite in the Type III rocks is very poor in Fe_2O_3, and ferric iron is scarcely calculated.

Equilibrium temperature and f_{O_2}

Text-fig. 5 shows log f_{O_2}-1000/T diagram estimated by the chemistry of coexisting Fe-Ti oxides. The calculating procedure follows the method of Spencer and Lindsley (1981) and Stormer (1983).

In the Type I rocks, bulk compositions of ilmeno-magnetite and hemo-ilmenite are used under the assumption of equilibrium of both minerals. This assumption, however, may not be adaptable to the monzogabbro, whose hemo-ilmenite is an interstitial mineral and is probably the product of the latest stage of the crystallization. The calculated temperature ranges from 750° to 850°C which indicates magmatic condition. The f_{O_2} is appreciably higher than the NNO buffer. In the case of the monzogabbro, this value may be meaningless. However, this rock probably crystallized under high f_{O_2} condition at least in the later stage, because its hemo-ilmenite shows appreciably high Fe_2O_3 content.

In the case of magnetite host and ilmenite lamellae of ilmeno-magnetite, calculated temperature and f_{O_2} indicate the equilibrium under subsolidus condition (Buddington and Lindsley, 1964). In addition, subsolidus equilibration is greatly affected by the modal ratio of magnetite and ilmenite (Morse, 1980). Therefore, f_{O_2} of subsolidus con-



Text-fig. 5 log f_{02}-1000/T diagram calculated by the method of Spencer and Lindsley (1981) and Stormer (1983).
Ind: calculated using the pair of independent ilmeno-magnetite and hemo-ilmenite, Comp: calculated using the pair of ilmenite lamellae and magnetite host of the composite grain. Other abbreviations are the same as in Table 2.

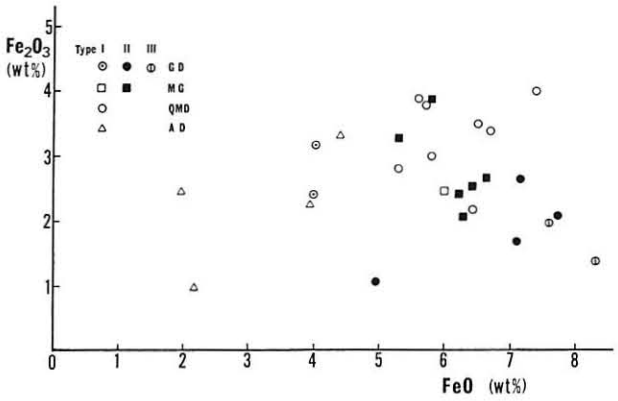
ditions occasionally shows a wider range than in magmatic conditions. Nevertheless, the f_{O_2} calculated by the composite grains in Type I is also high approaching the HM-buffer, and occasionally exceeds the valid limit of Spencer and Lindsley (1981). Hence, the Type I magma has probably crystallized at considerably high f_{O_2} conditions throughout magmatic to subsolidus temperature.

In the Type II rocks, calculated temperature and f_{O_2} show a wide range, but the f_{O_2} is lower than the Type I. Calculated temperature is rather low, less than 650°C even in the case of independent grains. Therefore, Fe-Ti oxides in the Type II rocks are considered to be appreciably reequilibrated at subsolidus conditions. The monzogabbro shows the highest temperature and f_{O_2} among the Type II rocks. The other Type II rocks plot near or lower than the QFM buffer. The specimen No. 52706, which consists of mixed facies of Types I and II, plots in a considerably wide area.

Since the Type III rocks contain no magnetite, temperature and f_{O_2} cannot be obtained by this method. The magnetite-series rocks, however, are considered to crystallize at higher f_{O_2} than the ilmenite-series ones (Shimazaki, 1976). In addition, the

chemistry of ilmenite in the Type III rocks is characterized by lower Fe_2O_3 than that in Type II, which suggests that the Type III magma was probably lower in f_{O_2} than the Type II magma. This is consistent with the investigation of Fe-Ti-O-S system, that is the ilmenite + pyrrhotite assemblage is stable at lower f_{O_2} than the ilmenite + magnetite + pyrite assemblage (e.g. Mariko et al., 1975; Nesbitt, 1982).

Text-fig. 6 shows an Fe_2O_3-FeO diagram of bulk rock composition except for the cumulates and aplites. The Fe_2O_3/FeO ratio of bulk rocks may indicate an oxidation state of magma (Tsusue and Ishihara, 1974). In the Matsumae plutonic rocks, the Type I shows the highest Fe_2O_3/FeO ratio, whereas the Type III the lowest. This fact coincides with the above estimation of f_{O_2}.



Text-fig 6 Fe_2O_3-FeO diagram of bulk rock composition except for the cumulates and aplites. Abbreviations are the same as in Table 2.

Discussion

The Matsumae plutonic rocks are divided into Am and P1 series, and the origin of the two series can be explained in terms of the difference in f_{H_2O} (Tsuchiya, in press). On the other hand, from the viewpoint of Fe-Ti oxide minerals, these rocks are divided into three types. It is noticed here that there is no strict correlation between assumed f_{H_2O} and f_{O_2}. The genesis of the three types is discussed below.

Czamanske et al. (1981) stated that the magnetite-series magma is generated either by oxidation at shallower level or intrinsically oxidized magma related to oxidized source materials. The distinction between them can be made by the mineralogy of the earliest stage of crystallization (Czamanske et al. 1981). In the case of the Matsumae plutonic rocks, the hornblendite II is considered to be the cumulate crystallized at the earliest stage judging from the high Cr_2O_3 content of clinopyroxene. Thus, the initial oxidation state of the Matsumae plutonic magma is probably represented by the hornblendite II, i.e. Type II. On the other hand, the Type I magma, which crystallized at higher f_{O_2} than the Type II magma, is therefore considered to have suffered some oxidation.

Czamanske et al. (1981) suggest that the oxidation trend of magnetite-series granitoids is brought about by separation of an aqueous phase and diffusive loss of H_2

through fractured roof rocks. In the case of the Matsumae plutonic rocks, diffusive loss of H_2 through country rocks may account for the oxidation of magma. The separation of an aqueous phase, however, may not be the main cause of the oxidation, because no correlation exists between degree of oxidation and expected depletion in Cl content of apatite (Tsuchiya, unpublished data).

As previously noted, the coarse grained (or slowly cooled) rocks of the monzogabbro show higher f_{O_2} than the fine grained (or rapidly cooled) ones. This fact probably indicates that the monzogabbro magma was oxidized after intrusion. On the other hand, the hornblendite I of Type I has subhedral to euhedral hemo-ilmenite, which suggests that some parts of the hornblendite I magma were oxidized at a relatively early stage of crystallization. This can be explained by the hypothesis that diffusive loss of H_2 occurred in the magma chamber crystallizing the hornblendite I. Such diffusive loss of H_2 is thought to occur in natural magmatic processes (e.g. Osborn, 1959; Sato, 1976).

Some rocks of the hornblendite I (e.g. No. 52706) show a mixed facies of Types I and II. This fact probably indicates that a less oxidized primitive magma (Type II) was incorporated into the oxidized magma chamber (Type I) and both magmas were mixed. This model of incorporation and mixing of primitive magma is consistent with the occurrence of Cr-diopside in some cumulates and complicated variation in Cr content of early-crystallized clinopyroxene in some rock types (Tsuchiya, in press). A similar model such as steady-state mixing in a magma chamber proposed by O'Hara (1977) has been emphasized recently in the case of calc-alkaline batholith (e.g. Barnes, 1983).

If the model of incorporation of less oxidized magma is adopted, the degree of oxidation in the magma chamber would reveal an extent of magma incorporation. Therefore, cessation of incorporation of magma would result in yielding of more oxidized and more differentiated magma.

In the case of the Am series of Matsumae, the earlier stage of crystallization is affected by magma incorporation as stated above. However, in the later stage, the Am series magma became more acidic (leucocratic granodiorite magma) by amphibole-dominant fractionation (Tsuchiya, in press). Since some rocks of the leucocratic granodiorite are regarded as Type I, the Am series magma probably oxidized at the later stage. This can be explained by the above model that oxidation and differentiation advanced when the magma incorporation ceased. On the other hand, the P1 series magma generally indicates high oxidation state. In addition, evolution of the P1 series magma can be clearly explained by a simple fractional crystallization (Tsuchiya, in press). This fact suggests that the incorporation and mixing of primitive magma was not so important in the P1 series.

The Type III rocks are characterized by the lowest f_{O_2} at crystallization among the Matsumae plutonic rocks. Their occurrence is restricted to a limited part of the melanocratic granodiorite, which possibly suggests local reducing conditions in the magma, e.g. interaction with carbon-bearing sediments. In this connection, other plutonic rocks in southwestern Hokkaido, which consist mainly of ilmenite-series rocks, have a possibility of larger scale of interaction with carbon-bearing sediments

(Ishihara, 1977; Shibata and Ishihara, 1979). Petrogenesis of these plutonic rocks in southwestern Hokkaido will be discussed in a separate paper.

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