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PHENOCRYSTIC SPINELIFEROUS TITANOMAGNETITES FROM  
TRACHYANDESITES, IKI ISLAND, JAPAN

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INTRODUCTION

Titanomagnetite is one of the most important rock forming minerals in basic igneous rocks; extensive studies of natural and synthetic iron ores have been made by many investigators over the past decade. These studies have furnished much data on the mineralogical character and petrological significance of titanomagnetites of igneous and metamorphic rocks.

Although theoretical and experimental relation indicates that magnetite ( $\text{Fe}_3\text{O}_4$ )-ulvöspinel ( $\text{Fe}_2\text{TiO}_4$ ) form a complete solid solution series with the spinel ( $\text{MgAl}_2\text{O}_4$ )-hercynite ( $\text{FeAl}_2\text{O}_4$ ) solid solution series at high temperatures, natural titanomagnetites that have crystallized as primary phases from basaltic magma have compositions on or near the join  $\text{Fe}_3\text{O}_4\text{-Fe}_2\text{TiO}_4$  in the system  $\text{TiO}_2\text{-FeO-Fe}_2\text{O}_3$ , and other components such as spinel-hercynite solid solutions and magnesiochromite-chromite solid solutions are generally negligible in titanomagnetites.

The present investigation deals with the titanomagnetite phenocrysts from trachyandesites of Iki Island, Japan and shows the extent to which a spinel-hercynite solid solution is included in titanomagnetites crystallized from an alkali basalt magma.

OCCURRENCE AND MICROSCOPIC OBSERVATION

The mineralogy and petrology of the alkaline volcanic rocks of the Iki Island region of southwestern Japan have been described elsewhere (Aoki, 1959, 1963a, 1963b, 1964).

TABLE 1. CHEMICAL ANALYSES OF TITANOMAGNETITES FROM IKI AND ADJACENT ISLANDS

	1	2	3	4	5	6	7	8
SiO <sub>2</sub>	0.23	0.51	0.03	0.28		0.32	0.26	1.16
TiO <sub>2</sub>	12.38	13.09	17.63	17.62	16.96	11.46	19.94	26.48
Al <sub>2</sub> O <sub>3</sub>	11.52	6.56	6.85	7.50		11.13	4.65	1.56
Fe <sub>2</sub> O <sub>3</sub>	41.10	45.30	43.47	30.71	39.78	45.25	26.77	36.21
Cr <sub>2</sub> O <sub>3</sub>	0.00			0.00			0.34	
V <sub>2</sub> O <sub>3</sub>	0.65	0.25		0.31		0.30	0.57	
FeO	30.03	29.92	29.13	39.55	34.27	27.97	44.89	31.08
MnO	0.28	0.44	0.56	0.52		0.34	0.76	1.04
MgO	3.90	3.77	2.33	3.02		4.52	2.18	1.31
CaO	0.03	0.15	0.04	0.15		0.11	0.03	1.07
<b>Total</b>	<b>100.12</b>	<b>99.99</b>	<b>100.04</b>	<b>99.66</b>	<b>91.01</b>	<b>101.40</b>	<b>100.39</b>	<b>99.91</b>
Mol. %								
MO <sub>1</sub>	49.3	50.3	45.7	56.5	50.8	48.3	59.6	46.3
R <sub>2</sub> O <sub>3</sub>	35.6	33.8	32.9	23.8	26.6	37.5	19.2	22.9
TO <sub>2</sub>	15.1	15.9	21.4	19.6	22.6	14.1	22.2	30.8
Spinel	23.7	15.2	15.6	15.8		22.6	10.0	
Ulvöspinel	33.4	40.8	51.2	48.5		30.7	55.4	
Magnetite	42.9	44.0	33.2	35.7		46.7	34.6	

<sup>1</sup> MO = FeO + MgO + MnO + CaO, R<sub>2</sub>O<sub>3</sub> = Fe<sub>2</sub>O<sub>3</sub> + Al<sub>2</sub>O<sub>3</sub> + V<sub>2</sub>O<sub>3</sub> + Cr<sub>2</sub>O<sub>3</sub>, TO<sub>2</sub> = TiO<sub>2</sub> + SiO<sub>2</sub>.

Phenocrystic titanomagnetites are rather common minerals in the trachyandesites but are rare in the mugearites in this area. In the former rocks, octahedral titanomagnetite phenocrysts commonly constitute about one wt. per cent, most are less than 5 mm in size, but some attain 20 mm when associated with very large kaersutite phenocrysts. The grain size and modal proportion of phenocrystic titanomagnetites gradually decrease as the host rocks diverge from trachyandesite toward both trachybasalts and trachytes. On the other hand, in the latter rocks titanomagnetite phenocrysts, idiomorphic or skeletal in form, amount to less than 0.1 per cent, and rarely exceed 0.3 mm in size.

Under reflected light the titanomagnetite displays no exceptional features. When observed in the polished sections, titanomagnetites Nos. 4 and 7 in Table 1 are homogeneous, but the latter mineral has rare weakly developed exsolution lamellae of ilmenite separated out along the (111) plane of the host minerals. Titanomagnetites Nos. 1, 2, 3 and 5 have alteration zones of various widths along crystal margins and cracks much like those described by Katsura and Kushiro (1961). In addition, they contain many fine ilmenite exsolution lamellae parallel to the (111) plane.

TABLE 2. FORMULAS OF TITANOMAGNETITES OF TABLE 1 ON THE BASIS OF 32 OXYGENS

	1	2	3	4	5	6	7	8
Si	0.062	0.142	0.008	0.080		0.085	0.074	
Ti	2.529	2.740	3.654	3.740	4.11	2.307	4.305	5.93
Al	3.690	2.151	2.228	2.497		3.514	1.575	
Fe <sup>'''</sup>	8.395	9.493	9.010	6.509	9.66	9.120	5.779	8.11
Cr	0.000			0.000			0.076	
V	0.141	0.057		0.036				
						0.064	0.131	
Fe <sup>''</sup>	6.815	6.968	6.708	9.316	9.23	6.264	10.768	7.73
Mn	0.064	0.104	0.131	0.124		0.077	0.185	0.26
Mg	1.593	1.576	0.965	1.270		1.804	0.941	
Ca	0.008	0.045	0.012	0.046		0.032	0.009	
XY	23.297	23.276	22.716	23.618	23.00	23.267	23.843	22.03

1. Phenocrysts of hypersthene bearing olivine titanaugite kaersutite trachyandesite, lava flow, Numazu, Iki island. Analyst, K. Aoki.

2. Phenocrysts of olivine titanaugite kaersutite trachyandesite, scoria fall, Numazu, Iki island. Analyst, K. Aoki.

3. Phenocrysts of olivine titanaugite kaersutite trachyandesite, scoria flow, Shofure, Iki island. Analyst, K. Aoki.

4. Phenocrysts of olivine titanaugite kaersutite trachyandesite, scoria flow, Shofure, Iki island. Analyst, K. Aoki.

5. Phenocrysts of trachyandesite, scoria flow, Shofure, Iki island. Analyst, T. Katsura (Aoki, 1959).

6. Phenocrysts of basaltic rock, Iki island. Analyst, S. Yamada (Matsui, 1958).

7. Phenocrysts of olivine titanaugite mugearite, laval flow, Madara island near Iki. Analyst, K. Aoki.

8. Groundmass of aphyric mugearite, lava flow, Madara island. Analyst, K. Aoki.

#### CHEMICAL COMPOSITION

Purification of titanomagnetites is one of the most difficult and tedious problems of mineral separation. Titanomagnetites from scoria falls and flows were picked by hand from the slightly weathered parts with which they occur. Next, they were crushed to -60 mesh between brass plates. On the other hand, titanomagnetite phenocrysts from lava flows were crushed to -60 mesh for subsequent mineral separation. Grains were then carefully separated by a weak hand magnet in water.

Further purification of the titanomagnetite was carried out by the method described by Basta (1960).

The chemical analyses of eight titanomagnetites from trachyandesites and mugearites, including six new analyses, are given in Table 1, and atomic ratios on the basis of O=32 are given in Table 2.

The chemical analyses are recalculated into 100 mol. per cent in terms

of  $\text{TO}_2$  ( $\text{TiO}_2 + \text{SiO}_2$ ),  $\text{MO}$  ( $\text{FeO} + \text{MgO} + \text{MnO} + \text{CaO}$ ), and  $\text{R}_2\text{O}_3$  ( $\text{Fe}_2\text{O}_3 + \text{Al}_2\text{O}_3 + \text{V}_2\text{O}_5 + \text{Cr}_2\text{O}_3$ ) and are plotted on the  $\text{TO}_2$ - $\text{MO}$ - $\text{R}_2\text{O}_3$  triangular diagram (Fig. 1). Other recently analyzed igneous titanomagnetites and titanomaghemites are included for comparison in this figure. As shown in Fig. 1, two of the titanomagnetites (Nos. 4, 7) plot near the  $\text{Fe}_3\text{O}_4$ - $\text{Fe}_2\text{TiO}_4$  binary join, while the others are plotted on or near the join  $\text{Fe}_3\text{O}_4$ - $\text{FeTiO}_3$ , because the latter are slightly oxidized and a part of their  $\text{Fe}^{2+}$  is changed to  $\text{Fe}^{3+}$ . Akimoto and Katsura (1959) and Katsura and Kushiro (1961) demonstrated that the ratio  $32 (\text{R} + \text{M} + \text{T}/\text{O})$  in the titanomagnetites from volcanic rocks ranges between 24 and 23, whereas that of the titanomaghemites ranges from 21.6 to 20.6. In the titanomagnetite phenocrysts from the Iki Island area this ratio is 23.8 to 22.7. Consequently these values agree with the titanomagnetites mentioned above.

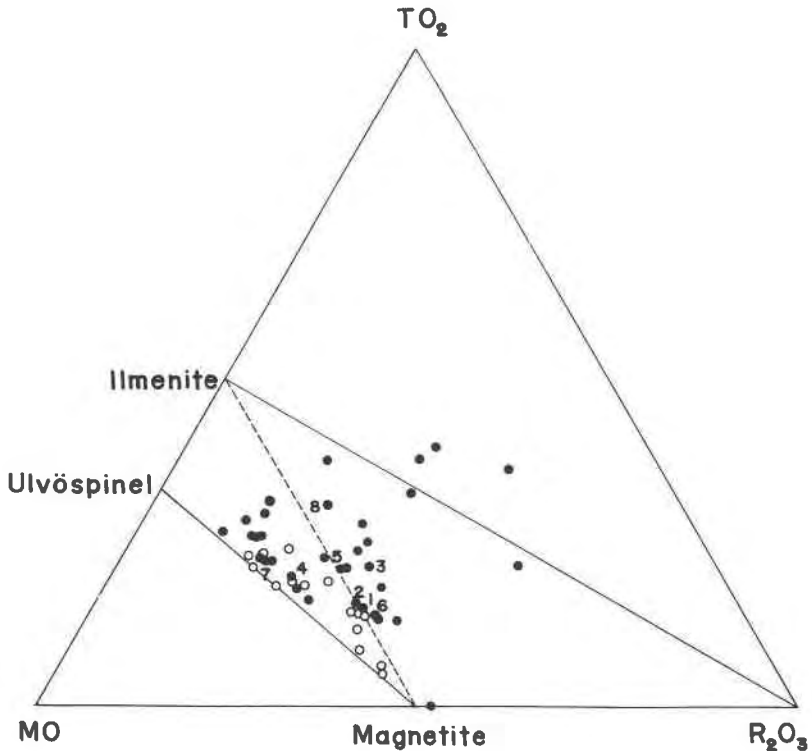


FIG. 1. Molecular composition of analyzed titanomagnetites.  $\text{MO} = \text{FeO} + \text{MgO} + \text{MnO} + \text{CaO}$ ,  $\text{TO}_2 = \text{TiO}_2 + \text{SiO}_2$ ,  $\text{R}_2\text{O}_3 = \text{Fe}_2\text{O}_3 + \text{Al}_2\text{O}_3 + \text{V}_2\text{O}_5 + \text{Cr}_2\text{O}_3$ . Solid circles: volcanic titanomagnetites and titanomaghemites, open circles: plutonic titanomagnetites. The numbers refer to analyses in Table 1.

The  $\text{TiO}_2$  and  $\text{V}_2\text{O}_3$  contents of the Iki titanomagnetites ranges from 12.4 to 17.6 and 0.3 to 0.7 per cent, respectively. They are normal values for igneous titanomagnetites.

#### SPINEL-HERCYNITE SOLID SOLUTION IN TITANOMAGNETITES

The titanomagnetite phenocrysts from trachyandesites of the Iki Island are characterized by high contents of  $\text{Al}_2\text{O}_3$  and  $\text{MgO}$ , ranging from 6.6 to 11.5 and 2.3 to 4.5 per cent, respectively. These abnormally high values suggest that spinel ( $\text{MgAl}_2\text{O}_4$ )-hercynite ( $\text{FeAl}_2\text{O}_4$ ) components are included as solid solution in the titanomagnetites.

In the magnetite-ulvöspinel series a continuous solid solution exists at high temperatures, with exsolution taking place below  $600^\circ\text{C}$ . (Kawai *et al.*, 1954; Vincent *et al.*, 1957; Wright, 1959; Basta, 1960). On the other hand, the relationships in the magnetite-hercynite series have been investigated by Turnock (1962) who indicated that complete solid solution exists above  $858^\circ\text{C}$ . Although the synthetic system spinel-hercynite has not been investigated, the study of natural minerals indicates that there is a continuous replacement series from spinel to hercynite (Deer *et al.*, 1962). From the high  $\text{MgO}$  and  $\text{Al}_2\text{O}_3$  contents cited above suggested that in the system magnetite-ulvöspinel- (spinel + hercynite) continuous solid solution exists at high magmatic temperatures.

In order to discuss the relative amount of components in analyzed magnetites, a calculation method for the normative mineralogical composition of magnetites was proposed by Vincent *et al.* (1957). This method is theoretically sound, but cannot be applied directly to volcanic titanomagnetites, which are usually too oxydized for analyses to reflect their original composition. The compositions of the titanomagnetites in Table 1 were calculated by a modification of the method of Vincent *et al.* It is first postulated that all titanomagnetites had compositions lying precisely on the  $\text{Fe}_3\text{O}_4$ - $\text{Fe}_2\text{TiO}_4$  join at some stage of crystallization. The chemical analyses of titanomagnetites were calculated into various spinel-structure components in the following order:  $\text{FeO}\cdot\text{Cr}_2\text{O}_3$  (chromite),  $\text{MgO}\cdot\text{Al}_2\text{O}_3$  (spinel),  $\text{FeO}\cdot\text{Al}_2\text{O}_3$  (hercynite),  $2\text{FeO}\cdot\text{TiO}_2$  (ulvöspinel),  $\text{MgO}\cdot\text{Fe}_2\text{O}_3$  (magnesian ferrite), and  $\text{FeO}\cdot\text{Fe}_2\text{O}_3$  (magnetite).  $\text{SiO}_2$  was added to  $\text{TiO}_2$ ,  $\text{MnO}$ ,  $\text{CaO}$  to  $\text{FeO}$ , and  $\text{V}_2\text{O}_3$  to  $\text{Fe}_2\text{O}_3$ .

The ratios of (spinel + hercynite):ulvöspinel:(magnetite + magnesian ferrite) calculated for titanomagnetites of the Iki Island area are shown in Fig. 2, together with those of other igneous titanomagnetites for which satisfactory analyses are available. Spinels from peridotite and gabbro inclusions of Iki Island are also included (unpublished analyses).

As shown in Fig. 2, it is clear that the plutonic igneous titanomagnetites include less than ten mol. per cent spinel + hercynite in solid

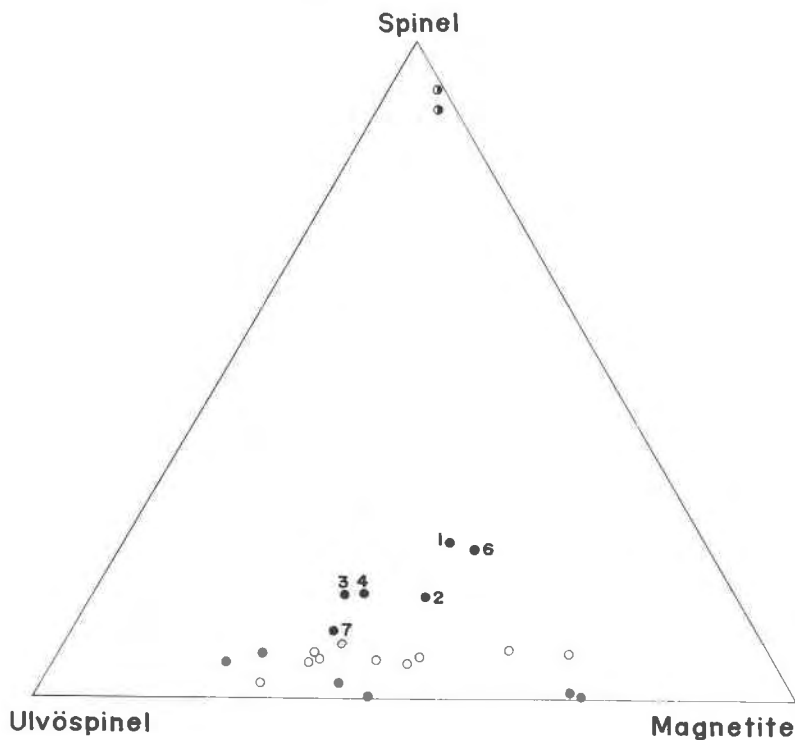


FIG. 2. Magnetite—ulvöspinel—spinel diagram. Magnetite =  $\text{Fe}_3\text{O}_4 + \text{MgFe}_2\text{O}_4$ , ulvöspinel =  $\text{Fe}_2\text{TiO}_4$ , spinel =  $\text{MgAl}_2\text{O}_4 + \text{FeAl}_2\text{O}_4$ . Symbols and numbers refer to Fig. 1.

solution. The titanomagnetite phenocrysts from Iki Island contain 15 to 24 per cent of spinel+hercynite components, the highest reported for natural titanomagnetites. It is suggested that the possible extent of solid solution in natural titanomagnetite in that area is at least 25 per cent spinel+hercynite.

It is not clear why such titanomagnetites with high spinel-hercynite solid solution have crystallized from the Iki trachyandesite magma. The fact that spineliferous titanomagnetite coexists with usually large kaersutite crystals and augite having a high  $\text{CaAl}_2\text{SiO}_6$  molecule content (Aoki, 1963a, 1964) seems to indicate that the trachyandesite magma crystallized at higher water vapor pressures and lower temperatures than did the mugearite magma.

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## CELL DIMENSIONS AND SPACE GROUP OF TAMARUGITE

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Tamarugite,  $\text{NaAl}(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$ , is a secondary mineral formed from the oxidation of sulfides, usually under arid conditions. Optical examination of crystals found with sideronatrite in a sample from Mina de la Compania, Sierra Gorda, Chile (U. S. National Museum, Smithsonian Cat. No. R6287) revealed  $\alpha = 1.485$ ,  $\beta = 1.487$  and  $\gamma = 1.498$ , all  $\pm 0.002$ . These thus agree with the values,  $\alpha = 1.484$ ,  $\beta = 1.486$ ,  $\gamma = 1.497$ , all  $\pm 0.001$ —reported by Gordon (1940). Similarly, the morphological and physical properties observed in the tamarugite of this present study confirm those reported by Gordon (1940). Published x-ray data on tamarugite were not found by the writers. Thus it was necessary to determine