

# THE ORIGIN OF CHROMITES OF THE TARI DISTRICT, TOTTORI PREFECTURE, JAPAN\*

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北原 順一：鳥取県多里地方産クロム鉄鉱の成因

## INTRODUCTION

The writer has studied the chromites in the Tari district, Tottori Prefecture and some facts relating to the minerals. The reports are included chiefly in the *Nihon Ganseki Kōbutsu Koshō Gakkai Shi* (The Journal of the Japanese Association of Mineralogists, Petrologists and Economic Geologists) and the *Shimane Daigaku Ronshū* (Bulletin of the Shimane University (Natural Science)). The author describes chiefly the origin of chromites in the present paper.

## RELATION AMONG GRANITIC AND ULTRABASIC ROCKS

The chronological relation of the intrusions of granitic and ultrabasic rocks was previously unknown because no data was found which served to solve the problem. As mentioned already granite porphyry, however, has been found to have invaded an ultrabasic rock about 1 250 m south of Shuttate-yama (951.8 m high). The granite porphyry is a marginal facies of biotite granite and seems to be produced at low temperatures with some amounts of hot water. It is proved that the granite porphyry is not metamorphosed by the ultrabasic rock which juxtapose the granitic rock because phenocrysts of orthoclase or microcline contained in the porphyry are fresh. The index of refraction of the orthoclase or microcline,  $n = 1.527$ . As shown by the chemical analyses in the previous paper, saxonite metamorphosed by the granite porphyry is poor in  $H_2O_+$ . It is because enstatite in the saxonite was intensely altered into talc, at higher temperatures than serpentinization.

## DIFFERENTIAL DIKES

Gabbro is a dike although it appears to be allogene and like diorite it traverses the country rock and chromite body (Fig. 1). Namely gabbro and diorite are dikes produced from a single magma. Plagioclastic rocks consisting almost wholly of oligoclase with negligible amounts of hornblende, are uncommon rocks. They occur as differential

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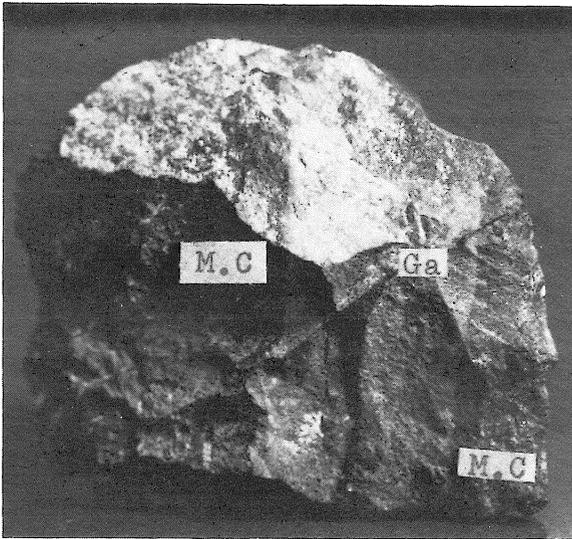


Fig. 1 Small gabbroic dike (Ga) intrudes in massive chromite (M.C).  
Deposit of Hirose mine

facies. Differential dike rocks have variable ratios between plagioclase and hornblende.

It seems to show that these dikes and dike-form inclusions belong to one series of igneous activity. Their inclusion began at or before the main stage of the plutonism and continued until the plutonism was completed.

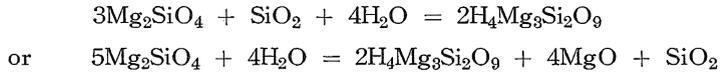
### PERIDOTITE AND SERPENTINITE

Serpentinite had intruded into palaeozoic rocks of older age and was also intruded by granitic rocks of younger age.

The Shuttateyama serpentinite is 1.4 km wide and 3 km long and trends N10° W. It forks on the north, stretching a branch N10° E. The Inazumiyama serpentinite trends N70°–80°E and is 3.1 km long and 1.3 km wide. Every bodies of serpentinite are ellipsoidal. The ultrabasic rock of the district consists of peridotite, saxonite, and pyroxenite and is more or less serpentinized.

Peridotite consists chiefly of olivine, with which may be associated more or less pyroxene, generally enstatite, and subordinate grains of magnetite and chromite. The relative proportion of these minerals varies greatly in different portions of a rock mass. Peridotite, saxonite and pyroxenite appear to be products of differentiation during the solidification of same original magma. The most common type in Tari district is saxonite, a rock rich in olivine with a considerable quantity of enstatite. Olivine and enstatite both alter to serpentine, but the enstatite generally alter less readily than olivine. Many dustlike particles and grains of magnetite are present in the serpentine and frequently show an irregular meshlike or gratelike arrangement determined by the lines of fracture in the altering olivine and enstatite, from which the magnetite in serpentine is chiefly derived. Chromite grains have a generally brown color or opaque by transmitted light.

Ferner and Verhoogen show that the serpentinization of olivine can be represented by following equations :



Of those equations the second may be expected to represent more closely the actual process. Serpentinized dunite may be formed by metasomatic or deuteritic alteration involving desilication of basic rocks by water vapour which is undersaturated with  $\text{SiO}_2$ . The most significant changes are loss of  $\text{MgO}$  and  $\text{SiO}_2$ , addition of  $\text{H}_2\text{O}$  and change of ferrous to ferric iron in the altered rocks. Forsterite reacts to give serpentine at about  $500^\circ\text{C}$ . However, if water is not available, forsterite is stable below  $500^\circ\text{C}$ , and in the absence of water it can form below  $400^\circ\text{C}$  magnetite and quartz, as shown by Neeks.

The color of serpentinite is grayish green, greenish yellow, yellowish brown, brown, reddish brown, grayish black, dark black, etc, according to the character of the country rock and the intensity of serpentinization. Peridotite is more altered than saxonite, which turns to bastite but is not much metamorphosed. As a whole enstatite is proofer against metamorphism than olivine. Enstatite changes into chlorite when the former contains aluminium. The microscope reveals that olivine is embedded in enstatite with an ophitic structure which shows that the former crystallizes earlier than the latter. Therefore peridotite can be thought to be earlier than pyroxenite composed of enstatite. Serpentinization proceeds along the margin and cleavage of olivine. With the action the olivine grain is rounded and dusty magnetite increases. A black serpentinite is rich in dusty magnetite while yellowish green one includes small or no dusty magnetite. The dusty magnetite separates from olivine and assembles in a dark serpentinite, generally being absent in light green or yellow serpentinite close to chromite bodies. The dusty magnetite in serpentinite forms at the serpentinization of olivine according to the following equations :



As shown above oxygen is necessary in the formation of dusty magnetite at serpentinization.  $\text{Fe}_2\text{O}_3/\text{FeO}$  is greater than that in the original rock. Serpentinite rich in olivine grain is of dark color.

When the olivine that is poor in iron such as containing  $\text{Fo}_{96}$  is serpentinized, the dusty magnetite is not usually separated from the olivine, but remains in the olivine as antigorite. The rock which contains the olivine described above therefore becomes light green or light yellow at serpentinization. When a rock which contains  $\text{Fo}_{92-93}$  is serpentinized, a portion of iron in olivine is liberated as dusty magnetite.

Under the microscope a rock illustrates that a fresh euhedral enstatite is surrounded by chromium-bearing magnetite and that fine veinlets of enstatite and chromite cutting olivine occur in the same rock. Therefore it is clear that some enstatite is earlier than chromian-magnetite and other enstatite and chromite is later than olivine.

According to the measurement of the index of refraction of olivine in the serpentinite from the district with immersion method, the magnesium olivine ranges from  $\text{Fo}_{92}$  to  $\text{Fo}_{97}$ .

As the melting point of olivine is so high that the invading of peridotite magma is inconceivable. But the melting point of an ultrabasic rock such as pyroxenite and peri-

dotite falls when  $\text{CO}_2$  and  $\text{H}_2\text{O}$  are contained. The ultrabasic magma invades as a mobile magma, along weak line caused by the deformation of the earth crust although the magma contains small amount of solid phases. It is concluded that ultrabasic(peridotitic) rock which consists of olivine suspended in a liquid silicate melt phase and which is derived from gabbroic or basaltic magma, invades and consolidates along the weak line of the earth crust in an early stage. In magnesian olivine ( $\text{Mg}_2\text{SiO}_4$ ) and silica ( $\text{SiO}_2$ ) series, olivine crystallizes out earlier than enstatite without regard for the composition of the solution. They act upon each other. The microscope reveals that olivine crystallized earlier than enstatite. That an ultrabasic rock is rich in olivine in part and rich in enstatite in part is due to the order of crystallization, or to the heterogeneity of a silicate magma. As described already the portion rich in olivine solidifies earlier than one rich in enstatite, it is clear that pyroxenite or an altered pyroxenite forms a dike. Dioritic or leucocratic dike is deduced to have invaded at the last stage of the differentiation.

### CHARACTERISTICS OF CHROMITE DEPOSITS

The deposit occurs mostly in peridotite and somewhere in the metamorphosed serpentinite which is derived from saxonite. The chromite deposit generally is lenticular with the long axis coinciding with that of serpentinite and is massive or porphyritic. Segregated chromite bodies sometimes form a portion of the ultrabasic rock and some chromites are thought to have injected into a rock which is not completely consolidated yet. In those case the ore body gradually grades into the country rock. On the contrary it is considered to have separated from a chrom-bearing ultrabasic magma in depth and intruded itself into a previously consolidated ultrabasic rock along the fissures. The intrusion metamorphoses the adjoining country rock which has been partly serpentinitized.

As the silicate and chromite melt do not mix, chromium-bearing silicate magma separates into two unmixed molten magma. Olivine and enstatite concentrate and crystallize as peridotite, saxonite or pyroxinite from a hydrous chromium-bearing peridotite. The solidifying temperature of olivine and chromite is not constant, for the chemical composition of olivine and chromite are variable. Forsterite, fayalite, and enstatite melt respectively at  $1.890^\circ\text{C}$ ,  $1.205^\circ\text{C}$ , and  $1.557^\circ\text{C}$ . The crystallizing point is of course controlled not only by the melting point but the pressure, gas, water, etc. Because the chromite and silicate magmas of the district are presumed to contain water and carbonic acid gas, the olivine and chromite are inferred to solidify at the lower temperature.

The smooth surface of the boundary between an ore body and the country rock and the scratched side of a fissure along which chromite invades can be observed at excavated working of a mine. The direction of the fissure along which the ore occurs is nearly parallel to the length of serpentinite. The boundary between a chromite deposit and the country rock is mostly clear.

Some margin of porphyritic ore grades into the portion of the country rock that is disseminated with particles of chromite. Massive chromite precipitates from a melt that is rich in chromite and poor in silicates, and porphyritic or disseminated ore from

magma which contains chromite and fair amount of silicates, namely porphyritic ore are caused when chromite and olivine are closely mixed. The porphyritic ore consists of anhedral grains 0.5 mm to 3.0 mm in diameter. Some early chromites produce a striped or fluidal structure due to the drag of flowing magma. In the massive ore chromite closely occurs and the associated silicate minerals are few while in the porphyritic ore chromite is associated with a fair amount of silicate minerals.

### MICROSCOPICAL OBSERVATION OF CHROMITE

Under the microscope the grains of chromite scattered through the country rock are seen mostly to be brown or black and partly to be yellowish brown, reddish brown or brownish yellow with a black margin. The disseminated ore is usually formed near the margin of the country rock; and a massive ore, far from that.

A microscope reveals that a chromite is traversed by irregularly crossed or curved and brown or black lines. Some is black and veinletlike. The margin of a porphyritic or disseminated ore is mostly black. The brown or black material is found also in a chromite from the other district, but its origin is unknown. Some consider that it is produced by a magma and other thinks that it is formed by the addition of iron when olivine is serpentinized.

A massive ore is generally seen to be brownish yellow or reddish yellow or traversed by an irregular brown latticework or brownish black straight or curved lines. A brownish or reddish yellow substance is high in a spinel composition. A brown one contains spinel and a little magnetite composition. Black one is rich in a magnetite composition. Cracks are usually found in a massive ore.

A shrinkage fissure in a chromite is filled with silicate minerals such as serpentine. The chromite is probably cracked when the adjacent olivine is expanded at serpentinization.

A silicate mineral such as serpentine cuts across the black line in chromite or fills the boundary between black margins. A silicate mineral in chromite solidifies after chromite with silicate solute crystallizes. Therefore serpentine which is included in chromite occurs along a black line which is high in magnetite composition and considered

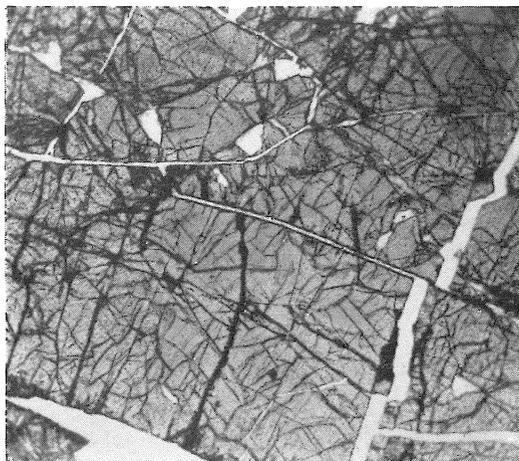
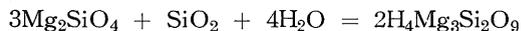


Fig.2 Massive chromite (sample No.6)  
Ogiri level of Hirose mine.  $\times 21$

to be younger than chromite. But silicate mineral is mostly included without relation with the black curved line (Fig. 2).

The serpentine is derived from olivine by the action of its own water. Serpentine does not generally associate with magnesite because the metamorphism is not caused by  $\text{CO}_2 + \text{H}_2\text{O}$  but by  $\text{SiO}_2 + \text{H}_2\text{O}$  as shown below.



The inclusion in a given chromite is the same with the substance which fills the space between grains. Thus in a sample from No. 7 level of Wakamatsu mine, for example, an inclusion in chromite grains are composed of olivine and enstatite. Here the olivine is earlier than the enstatite and produces a poikilitic texture. Some serpentine forming an inclusion is pierced by the other serpentine which fills the space between grains of chromite. Although their chemical composition is the same, the inclusion consolidates earlier.

The margin of disseminated ore usually is black. Yellowish brown chromite is traversed by brown lines or black irregular veinlets. Some chromite has a black margin, which sometimes extends to the inner portion. Silicate magma intrudes itself into fissures which are produced after the consolidation of chromite. In the sample a dark yellow portion is cut across by a brown lines and black irregular veinlets.

A microscope reveals that some chromite is varicolored and rich in dark brown or brownish black curved lines. For example, a sample from No. 7 level of Wakamatsu mine is traversed by brownish black spots or veinlets. Some marginal portions of the ore are black. Some section is black in part and other is nearly entirely black. Little ore is greenish yellow (hercynite  $\text{FeO} \cdot \text{Al}_2\text{O}_3$ ) and traversed with black irregular veinlet or skirted with a black substance. The brownish yellow chromite from the margin is traversed by a network of black lines and these increase gradually toward the surface of the ore body until the crystals completely replaced. Relics of the cherry brownish yellow chromite are in places preserved in the opaque black mineral.

A thin section of some ore is porphyritic in part, massive in part, and closely porphyritic in part. Mostly the massive ore is light brownish yellow and the porphyritic one is black with a brown portion. A massive ore is seen to be always associated with little olivines and serpentine derived from them, showing that chromite is of magmatic origin. Chromite is sometimes accompanied by talc, magnesite, chlorite, and chromic oxide garnet in some portion besides serpentine or antigorite.

When a brownish yellow or reddish yellow chromite which is rich in a spinel composition is crystallizing out,  $\text{Fe}_3\text{O}_4$  thus made is formed by the reaction of  $\text{FeO}$  and  $\text{CO}_2$  in hydrous magmatic emanation.  $\text{Fe}_3\text{O}_4$  thus made is metasomatically introduced to the margin of chromite which is crystallizing out, infiltrates fine interstices of the chromite, and metasomatically produces networks, which is rich in magnetite composition. Therefore the black margin and black crossing lines are closely related. The magmatic emanation is considered to contain substance which does not volatilize at the normal temperature.

When a small amount of magnetite composition is added to a chromite which has

been already solidified, a yellow portion turns in to a brown one. Consequently a brown margin becomes black. The process can be regarded as a metasomatism caused by magmatic emanation.

The cracks which are produced during the solidification of chromite is filled with residual magma or silicate melt. At the time a substance with magnetite composition or veinlets of magnetite are cut with residual magma in some place. Margins and sides of very narrow openings of chromite are corroded by a silicate melt, but chromite is not colored in black as the result of the corrosion. The margin of chromite is not colored black at serpentinization because the margin of chromite is already black when fresh enstatite comes in touch with chromite.

The country rock through which grains of porphyritic chromite or ore are scattered like phenocrysts through a groundmass is usually light green or light yellowish green because iron is metasomatically introduced to the chromite grains from the melt, silicate mineral lacks in iron, and the country rock is serpentinized. The iron content of the chromite grains is reverse to that of the surrounding silicate minerals. This is due to the above mentioned fact.

The massive ore is very slightly associated with silicate minerals that are formed at the same time. Therefore there is no quantitative relation between them.

The microscope reveals that the massive ore is less intensely influenced by magmatic emanation than a porphyritic ore. Consequently the iron content of a massive ore is generally less than that of a porphyritic ore.

Some closely porphyritic chromite includes olivine together with enstatite. Olivine and enstatite have in turn a chromite inclusion. Here the opening between chromite grains is filled with olivine and enstatite. Enstatite sometimes contains an olivine inclusion. Nearly all the olivines just mentioned are serpentinized and some of them are included by enstatite. The occurrence indicates that the chemical composition of the inclusion and that of the filling are the same.

Most chromites macroscopically are black, but some from the Hirose level of the Hirose mine is brown. The brown chromite usually is brittle and is seen, under the microscope, to be cracked. The brown color is caused by an aggregation of cracked chromite grains.

Microscope reveals that a few chromite is bordered with a green chromic garnet of the ugrandite group or gray grossularite. Some chromite the edge of which is black and rich in a magnetite composition is skirted with a yellow andradite. These garnets are produced by the reaction of chromite and calcium-bearing silicate melt. The solubility of CaO is increased by CO<sub>2</sub>. These garnets turn into one other according to the quantity of Cr, Al, and Fe<sup>3</sup>. They can be expressed as follows. 3CaO. (Cr, Al, Fe<sup>3</sup>)<sub>2</sub>O<sub>3</sub>. 3SiO<sub>2</sub> (uvarovite), 3CaO. (Al, Cr, Fe<sup>3</sup>)<sub>2</sub>O<sub>3</sub>. 3SiO<sub>2</sub> (grossularite), and 3CaO. (Fe<sup>3</sup>, Al, Cr)<sub>2</sub>O<sub>3</sub>. 3SiO<sub>2</sub> (andradite).

Small quantity of Fe<sup>2</sup> and Mg are included in them. Of the elements that cannot be crystallized out from silicate melt, CO<sub>2</sub> and CaO make calcite and aragonite veins.

## CHROMITES UNDER THE REFLECTIVE MICROSCOPE

Under the reflective microscope chromites are seen to be gray. A very few portion is grayish white, short and linear or lenticular. As it is present without relation with silicate minerals, it seem to have been produced from separated solute. The white portion is either a chromite with abundant magnetite composition or magnetite with chromium. The white portion that is seen to have black crossed lines under transmitted light and seen to be uniformly gray under the reflective microscope, is chromite although it contains magnetite composition.

The reflective microscope reveals that some portion is grayish white and veinlike and that silicate mineral cuts grayish white lines or veinlets, which are rich in magnetite composition.

Although a silicate mineral appears to have replaced chromite and produced substance rich in grayish white magnetite, but really silicate mineral invades the crack or boundary rich in magnetite composition. Generally no reaction is produced when silicate mineral comes into contact with chromite.

As already mentioned grayish white substance is thought to be a result of metasomatal introduction of magnetite emanation except the portion which is thought to have been formed by separation in solution.

## CRACKS IN CHROMITE

When hydrous chromite melt accompanied by a small quantity of silicate residual magma intrudes itself into the cracks in an ultrabasic rock, it solidifies with little serpentized olivine and enstatite. The openings between chromite grains are filled with a secondary silicate mineral. The first crack is caused in chromite at its solidification, and is filled with silicate residual magma. The crystallized minerals are olivine and enstatite. They are more or less serpentized by the water contained in the residual magma.

After the silicate mineral fills the crack in and opening around the margin of the chromite, the second cracks are produced in the chromite and silicate minerals. Some cracks are continuous through the chromite and silicate minerals. Some ion dissolved out from the ultrabasic rock by ground water is precipitated as a constituent of a clay vein of the saponite or garnierite under a certain condition after serpentization. Some of the clay vein is accompanied by magnesite. Subsequently the 3rd crack is made. It is a shear fracture and contains no minerals.

All the fine cracks which were formed simultaneously with chromite before these cracks are metasomatically impregnated with magnetite composition, which are given off by a magmatic emanation. Consequently cracks are formed by four stages in the strict sense of the word.

## GEOCHEMISTRY OF CHROMITE

The chromite deposit occurs mostly in serpentinite which is derived from saxonite.

The deposit generally is lenticular with the long axis coinciding with that of serpentinite.

The writer wants to clarify the difference of the chemical compositions of chromites according to their occurrences. They are taken at present from the Ōgiri and the 34m levels, the Hirose mine; and from the 7th level, the Minami level, and the Chūgiri level, the Wakamatsu mine. It is impossible to measure the iron content of the original silicate minerals basing on the index of refraction of serpentinite because dusty magnetite is set free when olivine or other silicate minerals associated with chromite are serpentinitized.

#### **Chromite from the Ōgiri level, the Hirose mine**

The ore body on the Ōgiri level (745 m), the Hirose mine strikes  $N5^{\circ}-20^{\circ}W$ , and dips  $35^{\circ}-75^{\circ}W$ . The boundary between the ore and the country rock is very clear in a certain place while it is obscure and fingers in other place. It is presumed that the ore has invaded the consolidated ultrabasic rock in the former; and unconsolidated in the latter.

The chromite in the outer portion of the ore body is poorer  $Al_2O_3$  and spinel component and richer in iron ( $FeO+Fe_2O_3$ ) and magnetite component. Chromite in the outer portion is more intensely metamorphosed than that along the edge. As described already the metamorphism of chromite has no connection with serpentinitization. The difference may be due to the occurrence and mineral association. Chromites rich in  $Cr_2O_3$  came from the outer portion of the ore body, are nearly massive or closely porphyritic, and associated with more or less enstatite and the derived chlorite.

#### **34 m level, Hirose mine**

The chromite ore body on the 34 m level strikes  $N5^{\circ}-30^{\circ}W$ , dipping generally westward and rarely eastward. The ore mostly is massive to the naked eye.

Chromites near the inner portion of ore body contain small amount of  $FeO+Fe_2O_3$ . Consequently a magnetite component is scarce and a spinel component is rich. These chromites are, therefore, not intensely metamorphosed. Chromite in the outer portion include much iron with relatively large amounts of magnetite component. Consequently they are intensely metamorphosed. The chromite in the outer portion is generally small in a spinel component. The iron content of chromites which are taken from the edge of the ore body or the vicinity is slightly greater than that from the inner portion and slightly less than that in the outer portion. They are brownish black and more intensely metamorphosed than those from the inner portion.  $FeO+Fe_2O_3$  is inferred to have concentrated upon the chromite in the outer portion from the inner portion at low temperatures and under a high vapour pressure at later stages of the growth of chromite. The  $TiO_2$  content of every samples is scanty although those from the outer portion incline to be richer in  $TiO_2$  than those from the inner portion.

#### **Chromite from No. 7 level, Wakamatsu mine**

The ore deposit worked on No. 7 level (791 m) is 70 m long and 40 m wide, generally

strikes  $N10^{\circ}-20^{\circ}E$  and dips  $20^{\circ}-50^{\circ}W$ .

The chromite within a short distance is not always similar in chemical composition. Massive chromite tends to be richer in  $Cr_2O_3$  and poorer in  $FeO + Fe_2O_3$  than the neighboring porphyritic one.  $Cr_2O_3$  content in chromite does not regularly grade from the inner to the outer portion. The chromite in the inner portion contains relatively small amounts of iron or magnetite component and much spinel component and is weakly metamorphosed while reverse in the chromite in the outer portion. The composition of chromite that contains much  $Cr_2O_3$  and small  $Al_2O_3$  is slightly abnormal and is associated with enstatite and the derived chlorite in addition to serpentine. Some chromites are thought to have been altered by the addition of iron from gabbro.

#### **Minami level, Wakamatsu mine**

The ore body on the Minami level (785 m), Wakamatsu mine is 75 m long and 45 m wide and extends to No. 7 and Chūgiri levels.

A fault marks the boundary between the country rock and the western part of the ore body. The fault strikes northward, dips eastward at an angle of  $80^{\circ}$ , and extend to the Chūgiri level. Chromites which lie close to the fault, are of brown color, which seems to be the effect of crushing by the fault, for the color is similar to that of streak.

#### **Chūgiri level, Wakamatsu mine**

The lowest portion of No. 7 ore body is on the Chūgiri level (772 m) and 45 m long and 40 m wide.

Chromites in the outer portion, rich in iron and magnetite component and highly altered while chromites near the outer portion, contain relatively large amounts of iron although they are not so intensely metamorphosed as the chromite in outer portion. Chromite on the edge of ore body is a little poorer in iron and less highly altered than chromites in the outer portion. Chromite which occurs in a small veinlet on the edge of the ore body, is scanty in iron and weakly metamorphosed. It contains relatively great amount of spinel component and small amount of magnetite component. Chromite which is abundantly associated with enstatite and the derived chlorite, is relatively rich in  $Cr_2O_3$ . The  $Cr_2O_3$  does not always increase with approach to the border of the deposit.

### **RELATIONS BETWEEN CHROMITE AND ITS ASSOCIATED SILICATE MINERAL**

That some silicate minerals in the outer portion of chromite body is poor in iron is due to the fact that iron concentrates only upon chromite. Because crystallization in molten magma is complicated  $Al_2O_3$  is not always scarce only in the outer portion. It is plausible that the varieties and chemical compositions of silicate minerals and chromite associated with them are affected by their environments and conditions of growth. Similar tendency about the chemical composition exists between chromite and silicate

minerals, although the reverse is present according to the difference of the condition of growth. Iron ( $\text{FeO} + \text{Fe}_2\text{O}_3$ ) content of silicate minerals is small but is greater than that of silicate minerals that are associated with chromite in the inner portion. It is either that iron concentrates upon chromite than upon silicate or migrate to chromite from the silicate groundmass. As in outer portion chromite and silicate minerals are poor in  $\text{Al}_2\text{O}_3$  this outer portion is originally not abundant in  $\text{Al}_2\text{O}_3$ . Along the edge of ore body silicate minerals and chromites are rich in  $\text{Al}_2\text{O}_3$  and they are thought to have been produced in the environment that is rich in  $\text{Al}_2\text{O}_3$ . In the marginal portion silicate minerals are rich in  $\text{FeO} + \text{Fe}_2\text{O}_3$  and a part of  $\text{Al}_2\text{O}_3$  is considered to have migrated from the silicate groundmass to the chromite when it is consolidating. Silicate minerals and chromites are poor in iron in the central portion of the ore body. A porphyritic ore is generally richer in iron than a massive one and probably iron migrates from a silicate groundmass to chromite when a porphyritic ore is forming.

When porphyritic or disseminated ore is formed as mentioned above, iron is introduced to chromite grains by the magmatic emanation from the silicate groundmass and silicate minerals like olivine becomes poor in iron. Consequently olivine alters to light yellow or light green antigorite by the action of its own water. Under the microscope some chromite is seen to be skirted with a light yellow chlorite. Aluminium is thought to have been transformed from chromite to a silicate melt and chlorite is considered to have been yielded by the action of chromite and olivine or enstatite.

#### Cr/Fe IN CHROMITE

As iron concentrates in the latter stage of the developing of chromite, Cr/Fe tells that the ore body on the 34 level of Hirose mine is formed earlier than that on the Ōgiri level of the mine. Every ore bodies of No. 7 deposit of Wakamatsu mine are continuous and the maximums of Cr/Fe in the marginal portion are 2.6, 2.4, and 2.2. Therefore it is conceivable that the development of chromite ore bodies proceeds from the Chūgiri level to No. 7 level through the Minami one. The Mg/Fe ratio is determined by the quantity of  $\text{Al}_2\text{O}_3$  in the chromite whose  $\text{Cr}_2\text{O}_3$  content is not variable.

A chromite whose Cr/Fe is above 3/1 is used for metallurgy and a chromite whose Cr/Fe is below 3/1 is employed for refractories.

#### PHYSICAL PROPERTIES OF CHROMITES

A liquid consisting of a standard mixture of  $\text{AsBr}_3$ ,  $\text{As}_2\text{S}_3$ , and S with a different amounts of  $\text{CH}_3\text{I}$  is prepared for the measurement of the refractive index of chromite. The index of refraction of the liquid rectilinearly changes with concentration of  $\text{CH}_3\text{I}$ . The index of refraction of the yellow, cadmium yellow, and ochre portion of powdered chromite can be obtained, but that of black one cannot be measured as it is opaque. The index of refraction of the sample which is homogeneous without black portions represents that of the sample, while that of the sample with black spots is not representative. According to Winchell's mineralogy, the index of four extrem ends of spinel

series, spinel, magnesian chromite, ferrian chromite, and magnetite are respectively 1.715, 2.054 (2.00 Calc), 2.12, and 2.42. The chromite(Mg, Fe)Cr<sub>2</sub>O<sub>4</sub> which consists of equal quantity of magnesian chromite and ferrian chromite is 2.06 in its index refraction. Some thin section are different in color but the same in the index of refraction, because the spinel series are a solid solution of four components mentioned. The relationship of the color of thin section of chromite and its index of refraction is as follows.

Yellow .....	1.860
Yellowish orange yellow to orange yellow .....	1.862
Yellow ocher .....	1.863-1.864
Cadmium yellow .....	1.864
Reddish cadmium orange to cadmium orange .....	1.861-1.867
Yellow ocher .....	1.868-1.871
Brownish orange yellow .....	1.868
Brownish yellow to orange yellow (nearly) .....	1.869
Light brownish orange yellow, light brown to light brownish ocher .....	1.870±
Orange yellow (saturated) .....	1.872-1.874
Light brown, light brownish ocher, yellow (saturated), light brownish orange yellow, yellowish brown, orange brown to ocher ....	1.875±-1.878
Light reddish brown, light reddish orange yellow, brown, orange brown to light reddish ocher .....	1.882-1.892

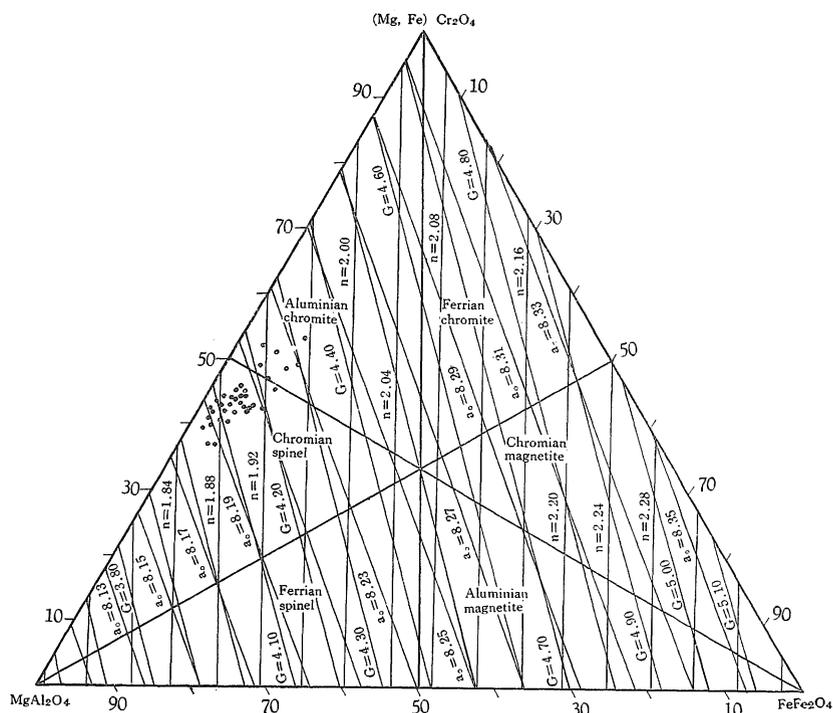


Fig. 3 Chemical classification and physical properties of chromite in Tari district. Each dot represents an analysis. By Kitahara

From the above description, it is ascertained that color of thin section with low index refraction is yellowish and red color increase with the rise of the index.

The lattice constant of spinel family are as follows : spinel 8.086Å, magnesian chromite 8.305Å, ferrian chromite 8.344Å, magnetite 8.374Å. Therefore the lattice constant of the chromite with  $\text{MgCr}_2\text{O}_4$  and  $\text{FeCr}_2\text{O}_4$  is 8.325Å.

The lattice constants of chromites from the levels are 8.188Å–8.224Å. That of the chromite from the marginal portion is greater than those from edge and the central portion. Correlation between the lattice constants and compositions of the chromites is apparent. Lattice constants are generally larger for porphyritic chromites than for massive chromites. The lattice constants are controlled by the FeO/MgO ratio, the larger constants being due to increase in the contents of the  $(\text{FeO} + \text{Fe}_2\text{O}_3)$  and  $\text{Cr}_2\text{O}_3$  compositions.

The four important terminals of the spinel group, i. e., spinel, magnesian chromite, ferrian chromite, and magnetite are respectively 3.55, 4.43, 5.09, and 5.20 in specific gravity. The chromite  $(\text{Mg, Fe})\text{Cr}_2\text{O}_4$  which consists of equal quantity of magnesian chromite and ferrian chromite is 4.76 in its specific gravity. The specific gravities of chromites from the levels of Hirose and Wakamatsu mines are 4.04–4.25 and 4.06–4.28 respectively. The mean specific gravity of 47 samples is 4.15. Specific gravities are generally larger for disseminated chromites than for massive ones. The larger specific gravities are due to increases in the magnetite component and/or decrease in the spinel component.

The chemical classification and the physical properties of chromite in the district are as shown in Fig. 3.

### ORIGIN OF CHROMITE

The studies on the origin of the chromites requires the investigation of the geology of the district, the country rock, and the differentiated dike because chromite is derived from a chromium-bearing ultrabasic magma or an olivine gabbroic magma. The examination of various rocks and dikes yield by the magmatic activity which is related to the origin of chromite, clarifies the character, occurrence, and forming epoch of chromite. The magma here consists of a liquid silicate melt phase and a number of solid phases such as suspended crystals of olivine, pyroxene, etc. Some considers that the body of ultrabasic rock or peridotite is an aggregate of olivine crystals which separates from a gabbroic magma and invades itself in a viscous state in the early stage of differentiation. Thus the description of the country rock and differentiated dikes is inseparably related to the studies on the origin of the chromites.

The chemical composition of chromite is variable according a sample. Microscope reveals many kinds of chromite. Specimen from a limited locality are different in chemical composition and microscopical appearance. It is clear that chromite melt is not homogeneous. A body of ultrabasic rock consists essentially of olivine or enstatite or both olivine and enstatite. Serpentinization is different according to a portion. It indicates that magma is heterogeneous and the order of crystallization of minerals is not the same.

Chromite is always associated with silicate minerals. Some clear ore appears to be free from silicate with naked eye, but it contains microscopical silicate minerals. Namely chromite melt is not completely separated from chromium-bearing ultrabasic magma; it is always accompanied by silicate. According to the quantity of associated minerals, a chromite ore is various; one is porphyritic (disseminated) when it has a little chromite grains, some is porphyritic with many chromite grains, and another is massive and rich in chromite. At differentiation of a chromium-bearing ultrabasic magma, the constituents do not completely separate as described already; a little silicate enters into chromite while a small quantity of chromite is left in silicate magma.

An ultrabasic magma rich in  $\text{Cr}_2\text{O}_3$  precipitates chromite as there is no suitable chromium-bearing mineral except it. Hence  $\text{Cr}_2\text{O}_3$  combines with elements which are most attractive to it under the physicochemical conditions, forming a proper member of the chromite family. Thereupon the composition of the ultrabasic rock decreases by a certain quantities of  $\text{Fe}^2$ , Mg, Al,  $\text{Fe}^3$  which are necessary to form chromite with  $\text{Cr}_2\text{O}_3$ . Consequently chromite and the ultrabasic rock are inferred to be of the same origin and different phase. The FeO in the magma tends to combine with  $\text{Cr}_2\text{O}_3$  if present, in preference to MgO. This goes on until the  $\text{Cr}_2\text{O}_3$  is consumed, then FeO combines with MgO to form silicates, i. e., the presence of  $\text{Cr}_2\text{O}_3$  deprives the magma of FeO so that the silicates formed after that will be poor in FeO. This was determined by comparing the relative amount of the FeO and MgO in the chromites and associated silicate minerals.

Basic magma is more differentiable than acidic one. The chromium-bearing ultrabasic magma in the region separates into a molten material consisting chiefly of chromite and a magma consisting chiefly of a component of a ultrabasic rock. Magmatically segregated ore bodies form a portion of the igneous mass, and usually grades off into it, but in some cases the chromites have not only become differentiated from the parent magma, but this separated portion has been forced up from below, independent of the rest of the igneous mass, thus forming a true intruded mass. The distinction of the outline varieties from abrupt change to perfect gradation. There is sometimes evidence of alteration by the chromite, of displacement, or of alteration subsequent to formation of chromite. Away from the chromite, residual olivine is abundant, but serpentinization increases to completion as the chromite is approached.

The segregation must be supposed to have taken place in the magma chamber, the some molten chromite and peridotite being injected contemporaneously. Many a large part of the chromite of some ore deposits crystallized after first formed silicates of host rock. Jun Suzuki showed that chromite crystallized in the orthomagmatic stage, these chromite crystals were intruded into a weak zone of an ultrabasic rock as crystal mush from a deep seated chromite reservoir in the hydrothermal stage, and the veins and/or massive bodies of chromites were formed.

Chromite and ultrabasic rock are not perfectly separated; there is no clean chromite which is not mixed with more or less silicate minerals and no silicate rocks which does not include accessory chromite. The porphyritic chromite is like that of a igneous rock. The

ore body and the country rock contain the same minerals, but the relative abundance of the silicates and metallic minerals is reversed. The difference is only in the proportion of the amount of silicate minerals to that of chromite.

The chromite in the inner portion of the ore body is presumed to have been formed earlier than that in outer portion. From chemical analyses it is ascertained that the chromite in the inner portion consists chiefly of chromian spinel or picotite while that in the outer portion is composed mostly of the chromian spinel and aluminian chromite which are rich in the magnetite component, because they are crisscrossed and skirted by a black mineral. When chromite concentrates upon the inner portion and the chromian spinel or picotite which are high at a melting point begin to coagulate in the inner portion, volatile substances from magma such as water become abundant and the water vapor pressure increases. Consequently volatile matter concentrates upon the outer portion and crystallization proceeds from the inner portion to the outer portion. This phenomena is clarified by Yoder's research.

As forementioned chromite in the outer portion is crisscrossed and skirted with black material rich in a magnetite component by primary autometamorphism at the later stage of crystallization. The chromite becomes, as a whole, chromian spinel or aluminian chromite rich in magnetite component. This primary autometamorphism can be explain as follows. FeO is partly changed into Fe<sub>3</sub>O<sub>4</sub> by the effect of volatile matters given off by a magma at the later stage of crystallization of chromite, e. g., water and carbon dioxide and their heat. In addition Fe<sub>2</sub>O<sub>3</sub> combine with surplus FeO to form Fe<sub>3</sub>O<sub>4</sub>. As oxidation of iron is influenced by the concentration of water vapor and carbon dioxide from magma and the vapor pressure during crystallization, volatile matter is the cause of oxidation. The state of oxidation of the iron is directly controlled by the concentration or pressure of water in the melt, resulting in a high Fe<sub>2</sub>O<sub>3</sub>/FeO ratio. The core of chromian spinel or aluminian chromite components sometimes remain, but sometimes they are completely replaced with magnetite component. As Fe<sup>2+</sup> ion is larger than Mg ion, the melting point of Fe<sup>2+</sup> compound is lower than that of corresponding Mg ion. In the cooling residual magma FeO/MgO relatively increases and Fe<sub>2</sub>O<sub>3</sub> concentrates. Therefore the magma contains enough Fe<sup>2+</sup> and Mg to prevent the growth of Fe<sub>2</sub>O<sub>3</sub>. That when the cooling is gradual, differentiation proceeds well and that the physical conditions during cooling strongly affect the character of mineral are well known. As various chromites are produced by complicated reaction in melting state, chromite with Fe<sub>3</sub>O<sub>4</sub> component cannot be produced by a simple mechanism and reaction. That from mobile silicate magma which corresponds to groundmass migrate some substance (chiefly Fe<sub>2</sub>O<sub>3</sub>) to consolidating chromite and that other substances (chiefly Al<sub>2</sub>O<sub>3</sub>) migrate to silicate magma from chromite are plausible. The view is suggested by Bumpei Yoshiki's research. The chemical compositions of chromite and silicate groundmass are presumed to have been altered during developing. Olivine is thought to produce Fe<sub>3</sub>O<sub>4</sub> under hydrothermal reaction, which is as follows.



During the process enstatite and clinoenstatite are formed and Fe<sub>3</sub>O<sub>4</sub> may migrate to chromite by the effect of residual water vapour. That outer portion of ore body is rich

in magnetite component and is accompanied by enstatite may due to the mechanism just mentioned.

As a whole the massive ore associated with a small amounts of silicate mineral is abundant in the inner portion while the porphyritic one associated with silicate minerals is rich in the outer portion, but the reverse is found in a few places. Where magma separates into a magma consisting chiefly of silicate minerals and that consisting mostly of oxide, the former comprises a great amounts of olivine and the derived serpentine and subordinate amounts of enstatite and the derived chlorite while if the latter contains  $\text{Cr}_2\text{O}_3$  and sufficient amount of  $\text{MgO}$ ,  $\text{Al}_2\text{O}_3$ , the chromite that crystallizes at high temperatures in the inner portion of the deposit, contains much spinel ( $\text{MgO} \cdot \text{Al}_2\text{O}_3$ ) component poor in iron, i. e., chromian spinel or picotite. Their microscopical color varies from yellow through cadmium yellow to ochre with a slight change of the component. The serpentinized olivine which associates with them is rich in  $\text{MgO}$  and poor in  $\text{FeO}$ . As it is plausible that the melting point of  $\text{Mg}_2\text{SiO}_4$  is higher than that of  $\text{Fe}_2\text{SiO}_4$ ,  $\text{Mg}_2\text{SiO}_4$  crystallizes earlier than the  $\text{Fe}_2\text{SiO}_4$ . As  $\text{FeO} \cdot \text{Fe}_2\text{O}_3$  tends to concentrate on the cooling residual magma, chromian spinel or aluminian chromite rich in iron grows on the outer portion, the spinel component having a tendency to be richer in the inner portion than in the outer portion. Magnetite component crisscrosses or skirts chromite grain. The magnetic substance cutting irregularly across the chromite is not an ex-solution phenomenon, but rather minor replacement intersections. An iron riched chromite occurs around the border of, and along fissures in, the chromite grains. The magnetic chromite is plausible to have been formed through the migration of iron from silicate groundmass and/or the outward migration of iron from the centers of the chromite grains. Similarly with chromite, silicate minerals have a tendency to be richer in  $\text{FeO}$  toward margin than  $\text{MgO}$ . The associated silicate mineral is serpentinized olivine which is richer in iron in the outer portion than in the inner portion. Sometimes  $\text{SiO}_2$  is enriched in the residual magma. The  $\text{SiO}_2$  and the part of  $\text{Al}_2\text{O}_3$  that cannot enter into chromite form  $\text{Al}_2\text{O}_3$ -bearing enstatite or the derived chlorite. They associate with chromite. When magma is mobile substances can remove between chromite and silicate groundmass. It is plausible that  $\text{Al}_2\text{O}_3$  migrates to silicate groundmass and forms enstatite. A microscopical fluidal structure as well as a macroscopical porphyritic structure can be observed in the chromite in the outer portion. Such structure is considered to have been controlled by the conditions under which magma consolidated.

The view that the black rim of chromite is caused by the corrosive action of silicate magma or the absorption of iron at the serpentinization was definite. The black rim and black networks or crisscross are continuous. The corrosion theory cannot be advocated as a black substance is found in the sample that is too far from the margin to be attacked by a silicate magma. That iron does not migrate from a silicate groundmass to chromite at temperatures below  $500^\circ\text{C}$  at which serpentinization is believed to take place. Serpentinization is therefore not related to the alteration of chromite. The increase of volume of olivine at serpentinization cracks the adjacent chromite, which appears as if to have been penetrated by serpentine substance along crevices. Being cut across by a silicate mineral,

the black rim is not considered to have been formed by silicate magma. It is the product of a primary autometamorphism by the help of water vapor at the late stages of the deposition of chromite. The grains of disseminated chromite are enclosed in olivine which is partly altered to serpentine; the greater portion of chromite consolidates earlier than silicate minerals. The small irregular particles of original silicate included in chromite may have crystallized slowly, while the chromite proceeded more rapidly to completion, which was followed by the crystallization of most of the silicates.

Although  $\text{MgO} \cdot \text{Al}_2\text{O}_3$  and  $\text{FeO} \cdot \text{Al}_2\text{O}_3$  form a continuous series of solid solution, but only a limited quantity of  $(\text{Mg, Fe}) \text{O} \cdot \text{Fe}_2\text{O}_3$  can enter into the series. Therefore formation of homogeneous chromite is nearly impossible. Iron concentrates and black portion rich in magnetite component is produced in the primary autometamorphism by water vapor and its pressure at the later stage of crystallization. In a sample chromite rich in a spinel component and that rich in a magnetite component form respective solid solutions and show the difficulty of producing a homogeneous chromite.

The inner portion of the ore body is generally rich in a spinel component and poor in a magnetite component. The tendency is however not universe in a chromite ore. Some ore bodies are rich in spinel component in the outer portion. In the deposit just mentioned the amount of  $\text{Cr}_2\text{O}_3$  is poor even in the marginal portion. That magnetite is more scarce in the inner portion than the outer portion is always observed except the chromite body whose inner portion is always metamorphosed by gabbro. Chromite rich in  $\text{Cr}_2\text{O}_3$  is found in the outer portion. It is noteworthy that enstatite and the derived chlorite are associated with chromite rich in  $\text{Cr}_2\text{O}_3$ . Cameron and Emerson state that they cannot see why chromite in pyroxenite is rich in  $\text{Cr}_2\text{O}_3$ . Chromite rich in  $\text{Cr}_2\text{O}_3$  is relatively poor in  $\text{Al}_2\text{O}_3$ . Because chromite poor in  $\text{Al}_2\text{O}_3$  is associated with  $\text{Al}_2\text{O}_3$ -bearing minerals, enstatite or the derived chlorite, surplus  $\text{Al}_2\text{O}_3$  in chromite migrates to silicate groundmass. Consequently chromite rich in  $\text{Cr}_2\text{O}_3$  is associated with enstatite or the derived chlorite. Conversely chromite associated enstatite is not necessary in  $\text{Cr}_2\text{O}_3$ .

Chromites along the edge of ore body and in a veinlet which cuts across the country rock near the boundary of the main ore deposit are altered very little. They are inferred to have been produced from a mobile magma. They are thought to have been affected by water vapor or its pressure.

In short the chemical composition of chromite is influenced by associated minerals as well as surroundings. The effect of associated minerals is sometimes greater than the surroundings and vice versa in the other cases.

## SUMMARY AND CONCLUSIONS

In the present paper, the writer mainly described the origin on the chromites of the chromite bodies on the Ōgiri and 34 m levels of the Hirose mine, and the bodies on the 7th, Minami, and Chūgiri levels of the Wakamatsu mine.

Serpentinite is intruded into the paleozoic formation and invaded in turn by granitic rocks in the region. The writer's detailed research has clarified that gabbroic rocks were

dikes differentiated from an ultrabasic magma rather than xenoliths.

Segregated chromite forms a portion of the ultrabasic rock, and/or is metasomatically intruded into ultrabasic rocks before or after they consolidated.

The chromite in the district belongs to chromian spinel or aluminian chromite, which is rare in Japan as well as in foreign countries. The specific gravity of the chromite is 4.04-4.25, the index of refraction is 1.86-1.89, the lattice constant is 8.188A-8.224A.

Chromites in the inner portion of the chromite bodies on the above mentioned levels are not altered or slightly altered and poor in a magnetite component and rich in a spinel one. Chromites in the outer portion of the ore bodies are considerably altered and generally abundant in a magnetite component. Black borders and cross lines of the chromite are considered to have been formed from deuteric concentration of iron oxide generated by high pressure of magmatic emanation such as water vapor. Chromite is entirely replaced by the black substance in some portions of the surface of chromite. Chromite which forms the edge of a large body or a veinlet is not altered at all and relatively scanty in a magnetite component.

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