

Sulfide Minerals in the G and H Chromitite Zones of the Stillwater Complex, Montana

GEOLOGICAL SURVEY PROFESSIONAL PAPER 694



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By NORMAN J PAGE

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*The relationship of the amount,
relative abundance, and size of grains
of selected sulfide minerals to the
crystallization of a basaltic magma*



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SULFIDE MINERALS IN THE G AND H CHROMITITE ZONES OF THE STILLWATER COMPLEX, MONTANA

By NORMAN J PAGE

ABSTRACT

Pentlandite, pyrrhotite, chalcopyrite, niccolite, and platinum-group minerals occur in two chromitite zones (G and H) of the Stillwater Complex. Detailed mineralogic studies of stratigraphic sections of both zones illustrate that amount, relative abundance, and size of sulfide minerals are related directly to primary crystallization processes of a fractionating basaltic magma. Four textural categories of sulfide grains and aggregates are present: inclusions in cumulus chromite and olivine; grains associated with postcumulus phases; fracture fillings; and grains associated with alteration minerals. Averaging the sulfide minerals in the two zones, 2.5 percent occurs as inclusions, 88.4 percent occurs as grains associated with postcumulus phases, and 9.1 percent occurs as fracture fillings and in association with alteration minerals. Inclusions probably represent both immiscible sulfide droplets and crystalline sulfide minerals trapped by crystallizing cumulus olivine and chromite, whereas sulfide grains in postcumulus material represent interactions between sulfide droplets and the trapped magma. Temperature, partial pressures of oxygen and sulfur, and growth and settling rates of the sulfide and silicate phases were important factors in controlling the distribution of sulfide materials.

INTRODUCTION

Sulfide minerals have long been known to occur in the Stillwater Complex, southwestern Montana (Howland, 1933; Howland and others, 1936; Roby, 1949; Jackson and others, 1954; Peoples and others, 1954). The Stillwater Complex is a large tabular mass of mafic and ultramafic rocks of Precambrian age (Kistler and others, 1969) that crystallized in at least three distinct stages. The initial stage consisted of (1) the intrusion of basaltic magma into pelitic rocks, quartzites, and iron-formation—now metamorphosed to the pyroxene-hornfels facies—and (2) the crystallization of the Basal zone of the complex before the appearance of the first cumulus olivine (Hess, 1960; Jones and others, 1960; Jackson, 1961). The beginning of the second stage, deposition of the Ultramafic zone, is marked by the appearance of cumulus olivine. Jackson (1961) has divided the second stage into two in-

formal stratigraphic units—a lower Peridotite member and an upper Bronzite member. The Peridotite member contains about 15 cyclic units (Jackson, 1961, 1967, 1968) of which 12 or 13 contain chromitites. The third stage began with the appearance of the first cumulus plagioclase. During this stage, the Banded and Upper zones of the complex crystallized (Hess, 1960).

Sulfide minerals are concentrated in the Basal zone (Howland, 1933), in the chromitite zones of the Peridotite member of the Ultramafic zone (Page and Jackson, 1967), and in certain horizons of the Banded zone (Howland and others, 1936). Sulfide-rich pods and lenses, conformable with the igneous layering, occur at the base of the G and H chromitite zones (E. D. Jackson, oral commun., 1968).

The chromitite zones in the Stillwater Complex originated as direct crystallization products of a fractionating sheet of basaltic magma (Jackson, 1961, 1963, 1967, 1969), and the sulfide minerals, which are intimately associated with these zones, probably are comagmatic with the complex. Page and Jackson (1967) suggested that the sulfide minerals were localized either by a process of migration in fluids upward in the crystal pile, the sulfide minerals being trapped owing to the relative impermeability of the fine-grained chromitites, or by a mechanism of precipitation of mattes from the individual batches of magma that formed the cyclic units. This paper presents the available evidence concerning the relations of the sulfide, silicate, and oxide phase assemblages within two chromitite zones. These data can be used as limiting conditions on the mechanism of sulfide accumulation in the chromitites.

The descriptions of the sulfide mineralogy and petrology are based on samples collected by E. D. Jackson from the G and H chromitite zones (figs. 1, 2) in the Mountain View area of the Stillwater Complex. In this area the two chromitite zones are about 12 feet thick.

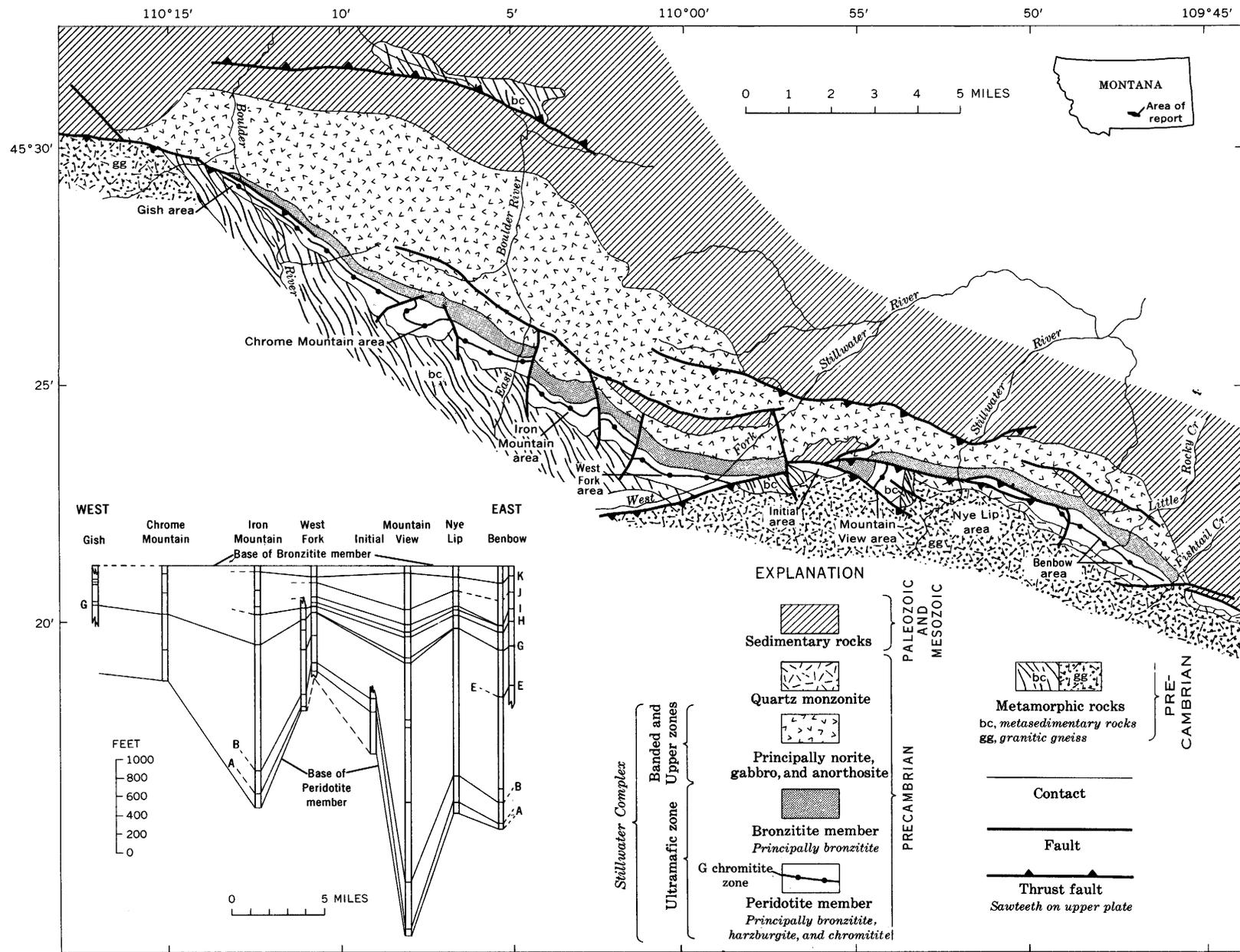


FIGURE 1.—Geologic index map and correlation diagram showing the chromitite zones in the Peridotite member of the Ultramafic zone of the Stillwater Complex. After Page and Jackson (1967).

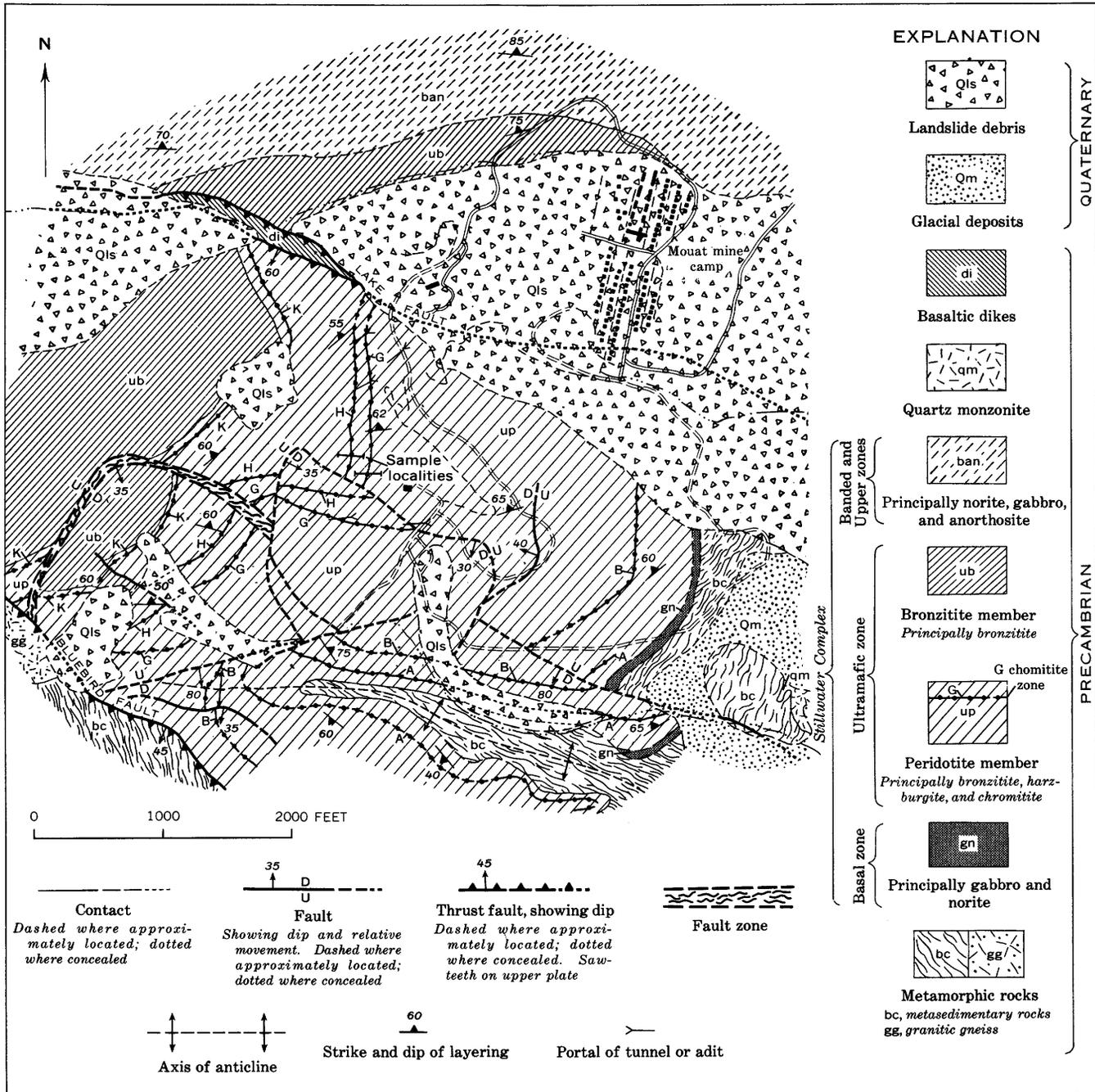


FIGURE 2.—Geologic map of the Mountain View area showing sample localities. After Jackson (1969).

Channel samples were cut across surface outcrops of the chromitite zones by Jackson (1969). Cores or chips, about 1 inch in diameter, were cut, mounted in epoxy, and polished to form sections suitable for electron microprobe investigations. Thirty-seven polished sections were prepared from the G zone, and 20 from the H zone. These polished sections were examined in reflected light, and thin sections from the same sample were examined in transmitted light. Each sulfide grain

was located in the polished section, and the following characteristics of the grain were cataloged: location, size (maximum and minimum outcrop diameter in microns), shape and textural characteristics, orientation with respect to stratigraphy (where possible), phases present, and boundary relations of the associated minerals. Photographs were taken for sample identification and in preparation for microprobe examination. Corrections were not applied to the measure-

ment of grain diameters in polished sections because the results are used for a comparative purpose within a single study. (See Jackson, 1961, p. 21, for further discussion.) Certain grains were X-rayed in powder cameras.

A large number of grains were studied (both qualitatively and quantitatively) with the electron microprobe, using metals and both synthetic and natural minerals as standards. Electron microprobe procedures were similar to those described by Page (1967) and Page, Calk, and Carr (1968), except that in the present study extensive use was made of X-ray beam scanning techniques and oscilloscope projection of the resultant images for a quick visual and qualitative identification of the elements present.

In order to obtain the volume percentages of sulfide minerals in the rock specimens, the sizes of the sulfide grains within a number of fields of view were tabulated, summed, and relative percentages calculated with respect to the total area of the fields of view that were examined in the section. The diameter of a single field of view was 778 microns, and on the average, 457.1 and 422.6 fields of view were examined per section in the G and H zones respectively. Polaroid photographs and a planimeter were used in estimating relative percentages of phases present in polymineralic grains. The precision error involved here is on the order of ± 20 percent and is based on replicate measurements.

ACKNOWLEDGMENTS

E. D. Jackson contributed the samples; he also shared knowledge of the silicate and oxide phases in the chromitite horizons and read earlier versions of this paper. The helpful and penetrating comments on earlier versions of this manuscript by A. L. Clark, G. K. Czamanske, G. A. Desborough, and P. B. Barton are warmly acknowledged.

SULFIDE OCCURRENCES

Within the G chromitite zone, a total area of 60.8 cm² was examined in which 1,816 sulfide grains were observed. In the H chromitite zone, a total area of 34.1 cm² was examined; 1,209 sulfide grains were counted. The H chromitite zone contains 0.01 percent sulfide minerals by volume, and the G contains 0.007 percent.

The range in diameter of sulfide grains in the G zone is from 1 to 185 microns (avg 5.5 μ), and the range in the H zone is from 1 to 135 microns (avg 11.1 μ). The average diameter of sulfide grains for the

G zone is strongly affected by the 1,413 grains of sulfide that have an average diameter of 1.9 microns. These grains are associated with alteration materials and thus may not represent a true value for the size of sulfide grains before alteration. If this size category is neglected, an average diameter of 18.1 microns is obtained, instead of 5.5, for the sulfide grains in the G zone.

Four textural¹ associations of sulfide mineral grains are found in the rocks of the G and H chromitite zones. These associations are: (1) inclusions of sulfide minerals, with or without silicate and oxide minerals, in cumulus olivine and chromite crystals; (2) sulfide minerals, with or without magnetite, associated with postcumulus minerals and interstitial to cumulus chromite and olivine; (3) sulfide minerals that form veins and fill fractures in cumulus phases; and (4) sulfide minerals that occur in veinlets formed by serpentinization and alteration of the primary silicate and oxide phases. Over 90 percent of the last two categories consist of pyrrhotite and pentlandite single-phase grains with an average diameter of 3.4 microns. Although they form 4.8 and 14.7 percent of the total volume of sulfide minerals in the G and H zones respectively, their occurrence marks them as probable postmagmatic features; therefore, they are not discussed further.

SULFIDE INCLUSIONS IN CUMULUS MINERALS

Pyrrhotite, pentlandite, chalcopyrite, niccolite, heazlewoodite, molybdenite, and minerals containing platinum-group metals, all with or without magnetite, phlogopitic mica, and other silicates, form inclusions usually within the centers of cumulus chromite and olivine crystals. The inclusions may contain one to three sulfide minerals in addition to silicates. These are the same types of inclusions discussed by Jackson (1961, 1966) and Page and Jackson (1967). The inclusions contain 1.4 percent and 3.1 percent of all the sulfides in the G and H zone chromitites respectively. The inclusions may be divided into the following four subcategories based on the cumulus host phase and the presence or absence of silicate phases within the inclusion:

1. Sulfide minerals that fill negative crystals, or sub-hedral sulfide crystals included in chromite.
2. Sulfide minerals of similar habit to 1 but included in olivine rather than chromite.

¹"Texture," as I use the term, includes the general fabric of the sulfide minerals in the rock and the grain sizes of these phases. The term "fabric" refers to the shape of the sulfides, their orientation, and the boundary relations between grains. The cumulus terminology is from Jackson (1967).

3. Sulfide minerals with silicate or oxide phases, or both, included as irregular negative crystals in chromite.
4. Sulfide minerals of similar habit to 3 but included in olivine rather than chromite.

The great majority of the inclusions (93.5 percent for the G zone and 91.0 percent for the H zone) occur in olivine. Inclusions in subcategories 3 and 4 have varying ratios of sulfide to silicate minerals, and these kinds of inclusions possibly represent a gradation between inclusions containing exclusively silicate or sulfide minerals.

FABRIC

Figure 3 illustrates the various shapes of minerals, types of contacts between the minerals, and kinds of sulfide minerals that occur as inclusions in the cumulus phases of the G and H chromitite zones. Where more than one mineral is present within an inclusion, euhedral negative crystals or subhedral forms are found, whereas, if a single sulfide mineral is present, the shape of the inclusion reflects a crystal form of the sulfide mineral. In stratigraphically oriented sections from the G chromitite zone, about 58 percent of the inclusions of subcategories 3 and 4 shows the sulfide minerals at the bottom of the inclusion.

Other specimens available from the G and H zones were oriented with respect to the layering, but not to the stratigraphy; for these specimens the stratigraphic top was assumed to be perpendicular to the layering. If a preferred orientation existed, the sulfide grains should be more abundant either toward the assumed top or toward the assumed bottom. There was a preferred orientation for about 76 percent of the inclusions in the H chromitite zone but not for the G chromitite zone.

Analyses of grain size data (averaged diameters) show that the average diameter of inclusions within the G and H zones is 4.9 and 6.7 microns respectively; the range of diameters for the inclusions in the G and H zones is from 1 to 35 and from 1 to 37.5 microns, respectively. Subcategories 1 and 2 on the average have smaller grain sizes than those of subcategories 3 and 4. In both the G and H zone chromitites, the size distributions are somewhat skewed toward the finer size classes and in general show moderate to well-sorted distributions. The sorting coefficients for the inclusions in the G and H chromitites are 1.43 and 1.60 respectively. The sulfide inclusions therefore give about the same value of sorting coefficients as Jackson (1961) found for crystals of cumulus chromite and olivine.

PHASE ASSEMBLAGES

The assemblages of sulfide minerals present in the inclusions are summarized in table 1. The term "abundance," as used here, refers to the number of

TABLE 1.—Abundance, in percent, of mineral assemblages occurring as inclusions

[Mineral symbols: pn, pentlandite; po, pyrrhotite; nc, niccolite; Pt, platinum-group minerals; ccp, chalcopyrite; mo, molybdenite; hz, heazlewoodite; mt, magnetite]

Mineral assemblage	Abundance (percent)									
	G chromitite zone					H chromitite zone				
	1	2	3	4	Total	1	2	3	4	Total
pn	3.3	11.0	4.4	6.6	25.3	5.0	23.3	5.8	13.3	47.5
po	3.3	2.2	3.3	6.6	15.4	5.8	.8	9.2	15.8	
nc	1.1	2.2	17.6	1.1	22.0	4.2	3.3	3.3	10.8	
Pt	16.5			1.1	17.6	6.7	3.3	.8		10.8
ccp		3.3	3.3	1.1	7.7	.8	1.7		.8	3.3
mo								.8		.8
pn+po		1.1			1.1		.8			.8
pn+ccp									.8	.8
pn+hz			1.1		1.1		.8		.8	1.7
pn+nc	1.1				1.1			.8		.8
pn+mt							.8		1.7	2.5
po+ccp				5.5	5.5				.8	.8
pn+phase Q				1.1	1.1					
po+mt				1.1	1.1		.8			.8
nc+mt							1.7			1.7
pn+po+Pt						.8				.8
pn+po+ccp				1.1	1.1					
Total	25.3	19.8	29.7	25.3	100.1	13.3	43.2	12.3	30.7	99.7

1 Sulfide minerals that fill negative crystals, or subhedral sulfide crystals included in chromite.

2 Sulfide minerals of similar habit to 1 but included in olivine rather than chromite.

3 Sulfide minerals with silicate or oxide phases, or both, included as irregular negative crystals in chromite.

4 Sulfide minerals of similar habit to 3 but included in olivine rather than chromite.

grains of the mineral or mineral assemblage compared to the total number of inclusions. The most common sulfide minerals present in inclusions in the G and H chromitite zones are pentlandite, pyrrhotite, niccolite, and the platinum-group minerals. Inclusions rarely contain more than one sulfide mineral. Platinum-group minerals are strongly concentrated as inclusions in chromite (subcategory 1), whereas pentlandite is concentrated as inclusions in olivine (subcategory 2). Except for these preferences, the sulfide minerals show no direct correlation with the inclusion subcategories.

SULFIDE MINERALS ASSOCIATED WITH POSTCUMULUS MATERIAL

Pentlandite, pyrrhotite, chalcopyrite, heazlewoodite, and complex platinum-group minerals occur in the chromitite zones interstitial to cumulus chromite and

olivine. Three subcategories were distinguished and catalogued for this type of material:

1. Sulfide grains which have sharp straight contacts with postcumulus phases and which are molded into all the available space between cumulus minerals.
2. Sulfide grains and grain aggregates that occupy interstices but have ragged, embayed contacts with the postcumulus phase and retain at least one grain boundary with the cumulus phases.
3. Sulfides with ragged, embayed, or (rarely) straight contacts entirely enclosed by postcumulus minerals.

Sulfide minerals associated with postcumulus minerals account for 93.8 percent of the total volume of sulfide minerals in the G zone and 83.0 percent in the H zone. In the G chromitite zone, 69.8, 24.1, and 6.0 percent of the grains are distributed between subcategories 1, 2, and 3 respectively, whereas in the H zone, 46.7, 37.4, and 15.8 percent of the grains are distributed in subcategories 1, 2, and 3 respectively.

FABRIC

Figure 4 illustrates the shapes and general appearance of a selection of grains in postcumulus interstices of the G and H chromitite zones. Most of the two-dimensional shapes are rectangular, triangular, and polyhedral forms which fill, or partially fill, the concave polygons of the interstices bounded by the crystal faces of chromite and olivine. The character of the interstitial spaces has been discussed by Jackson (1961, p. 65). Five field-oriented sections from the G chromitite zone were examined in detail; three of these appear in figure 4. In these sections, about 60 percent of the grains rests on the stratigraphic bottom of an interstice. In the remainder of the G zone sections, for which only the orientation on the layering plane is known, about 60 percent of the grains also rests on the stratigraphic bottom of an interstice. For the H chromitite zone, a maximum of about 59 percent of the grains rests on the bottom of the interstice if a stratigraphic top for the layering plane is assumed. No systematic distribution with respect to grain size, phases present, or subcategory has been found.

The sizes (outcrop diameter) of sulfide grains associated with postcumulus material range from 1.5 to 185 microns in the G chromite zone and from 1 to 135 microns in the H zone; the average size (mean diameter) is 26.5 microns for the G and 19.4 microns for the H. Subcategories 1, 2, and 3 have average diameters of 35.0, 32.0, and 13.9 microns respectively in the

G zone and 18.5, 23.5, and 14.1 microns in the H zone. Cumulative size distributions are skewed toward the finer grain sizes, and sorting coefficients are 2.25 for the G zone and 1.93 for the H; they indicate moderate to poor sorting.

Pentlandite, in single-phase or multiple-phase grains, was found in three ways: as subhedral to euhedral crystals without extensive fracturing and parting, as similar crystals that are highly fractured into mosaics of rhomb-shaped pieces, and as unfractured pentlandite grains with wormlike intergrowths of either pyrrhotite or phase Q, which is an unidentified yellow isotropic mineral or rarely, both in the same grain.

Pyrrhotite, in multiple-phase grains with pentlandite, was found as a wormlike intergrowth in which the worms of pyrrhotite are approximately 1 or 2 microns across; it may also be found as euhedral to subhedral crystals less than 5 microns wide that form a spongy-appearing aggregate. Pyrrhotite also was found as subhedral to anhedral irregular grains and as highly fractured grains in which the fractures are filled with magnetite. Boundaries between either the spongy or the subhedral to anhedral pyrrhotite and the pentlandite are usually straight and sharp and show no evidence of embayment or veining.

Chalcopyrite occurring with pentlandite or pyrrhotite has either sharp and straight contacts or forms veins and embayments into the other two minerals. Contacts between platinum-group minerals and other sulfides are usually sharp, and the shape of the contact is determined by the crystal form of the platinum-group mineral.

PHASE ASSEMBLAGES

Sulfide minerals and mineral assemblages filling or partially filling interstices between cumulus olivine and chromite are given in table 2, and their relative abundance can be compared for the G and H zones.

In figure 5, volume percentages of pentlandite, pyrrhotite, and chalcopyrite are plotted for a selected number of grains containing two- and three-phase associations of sulfides. Also included are some four-phase grains which contain magnetite as the fourth phase. Three-phase grains rarely contain more than 25 percent by volume of chalcopyrite and tend to have more pentlandite than pyrrhotite. Examination of the three-phase sulfide grains in the H zone suggests a weak stratigraphic variation from pentlandite-rich grains in the footwall to those rich in pyrrhotite in the hanging wall. Such a variation is partly supported by data from the G zone.

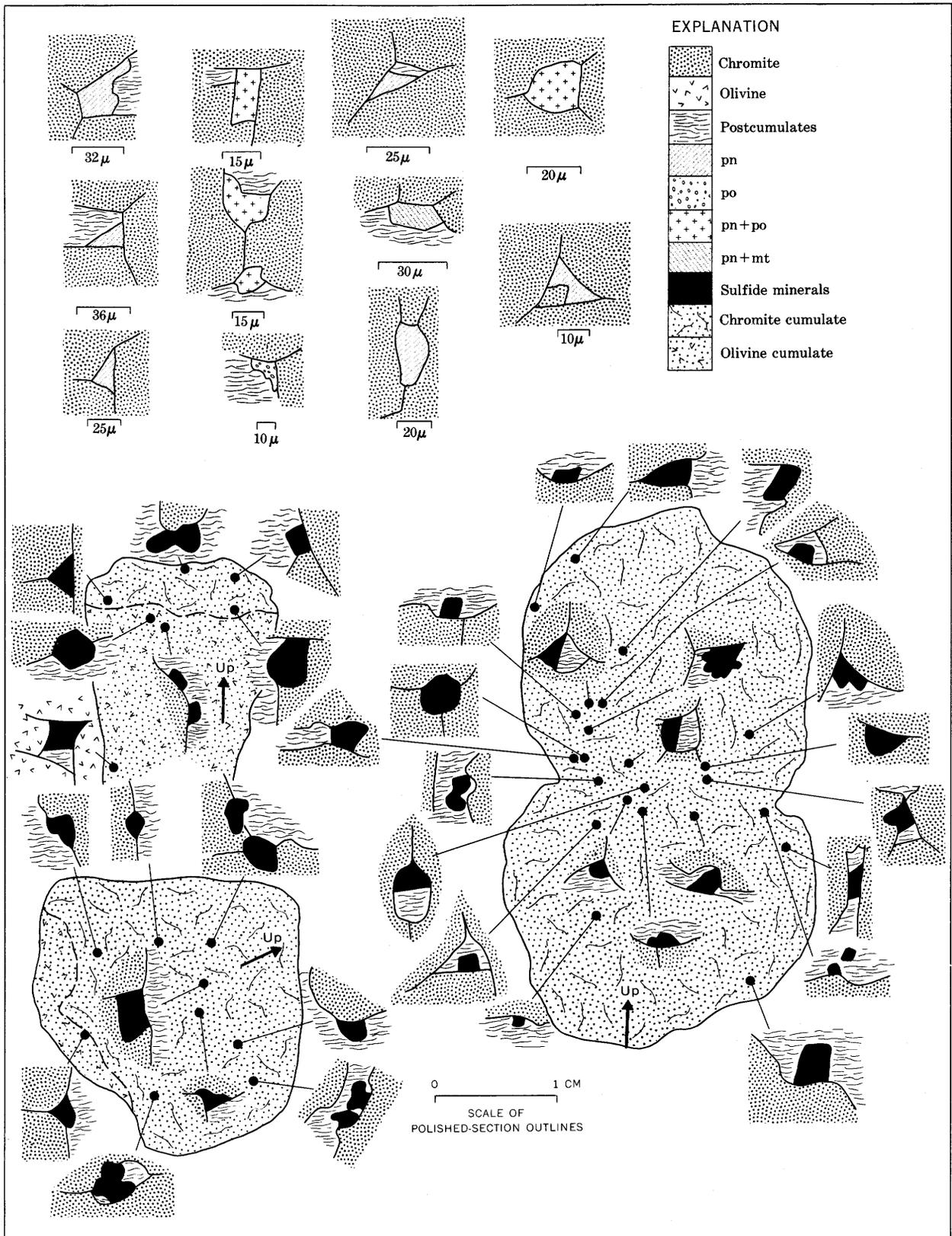


FIGURE 4.—Shapes of mineral grains associated with postcumulus minerals. Mineral symbols: pn, pentlandite; po, pyrrhotite, and mt, magnetite.

TABLE 2.—Abundance, in percent, of all mineral assemblages associated with postcumulus material¹

[Mineral symbols: pn, pentlandite; po, pyrrhotite; nc, niccolite; Pt, platinum-group minerals; ccp, chalcopyrite; mo, molybdenite; hz, heazlewoodite; vi, violarite; il, ilmenite; and mt, magnetite]

Mineral assemblage	Abundance (percent)		Mineral assemblage	Abundance (percent)	
	G chromitite zone	H chromitite zone		G chromitite zone	G chromitite zone
pn	43.8	52.2	pn+po+ccp	2.6	2.5
po	21.3	16.4	pn+hz+Pt	.4	.2
ccp	4.3	.9	pn+po+Pt	.4	---
vi	---	.2	pn+ccp+Pt	---	.2
nc	---	.3	pn+po+mt	3.4	.9
Pt	3.0	.2	pn+phase Q+mt	---	.3
pn+po	10.2	15.5	pn+hz+mt	.4	---
pn+spongy po	(3.8)	(10.0)	pn+Pt+mt	.4	.2
pn+po worms	(2.6)	(4.5)	po+phase R and S	---	.3
pn+spongy po+po worms	(3.8)	(1.0)	pn+po+phase Q+Pt	---	.2
pn+phase Q	.4	1.9	pn+po+phase Q+ccp	.4	---
pn+hz	---	.9	pn+po+phase Q+mt	---	.5
pn+ccp	1.3	.9	pn+po+ccp+vi	.4	---
pn+nc	1.3	.3	pn+po+ccp+Pt	---	.2
pn+mt	2.1	---	pn+po+nc+mt	---	.2
po+vi	---	.2	pn+po+ccp+mt	1.3	---
po+ccp	1.3	.9	pn+po+ccp+Pt+mt	---	.2
po+Pt	---	.2	1 phase grains	72.3	70.1
po+hz	.4	---	2 phase grains	18.7	23.1
po+mt	1.3	1.9	3 phase grains	6.8	5.5
ccp+mt	---	.3	4 phase grains	2.1	1.0
hz+mt	.4	---	5 phase grains	---	.2
pn+po+phase Q	---	.9			

¹ In the table certain phase assemblages have been grouped together. For the H zone, the abundance of pyrrhotite, 16.4 percent, includes 0.5 percent grains of pyrrhotitelike phase with an Ni to Fe ratio of 1 to 1. In the assemblage po+vi, the po represents a pyrrhotitelike phase with a Ni to Fe ratio of about 1 to 1. Phase Q, an un-

identified isotropic mineral with a strong yellow color, forms a wormy intergrowth with pentlandite. Phases R and S were found in only two grains and in amounts too small to identify, even with the microprobe. Phase R is blue gray in color, and phase S is a gold color.

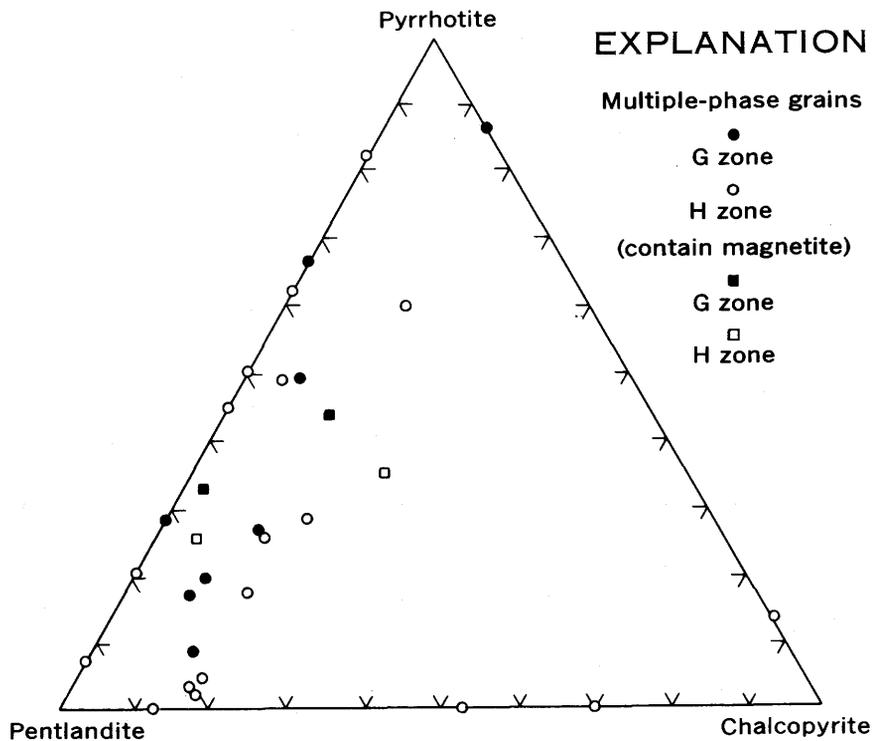


FIGURE 5.—Volume percentages of pyrrhotite, pentlandite, and chalcopyrite in multiphase grains.

Although pentlandite and pyrrhotite are the major sulfide minerals, both minerals are not present in all samples as interstitial material. There is no pentlandite in the olivine-chromite cumulate at the base of the G zone, and pyrrhotite is absent in the middle part of the G zone. In contrast, for the H zone, both pentlandite and pyrrhotite are present in every sample except those from the base; in the base samples only pyrrhotite is present. The distribution of sulfide grains in the upper part of the G zone is very similar to that of the whole H zone.

Examination of the sulfide grains and aggregates associated with postcumulus minerals shows that about 92.7 and 98.1 percent of all grains from the G and H zones respectively contain either pentlandite or pyrrhotite or both; about 43.8 and 52.2 percent of all grains consist of pentlandite, 21.3 and 16.5 percent of pyrrhotite, and 27.6 and 29.4 percent have both minerals. Other minerals, including chalcopyrite, niccolite, and the platinum-group minerals, constitute the remaining 7.3 and 1.9 percent of grains for the G and H zones, but these figures could be increased by possibly 2 or 3 percent by including the minor amounts of these minerals which are found in the complex aggregates of pyrrhotite and pentlandite. The distribution of one-, two-, three-, four-, and five-phase grains in the three subcategories is practically the same as the distribution of the same phases of grains on the average in both the G and H zones.

Sulfide grains associated with postcumulus material in the G and H zones have only 3.4 and 1.4 percent of platinum-group minerals and 0.0 and 0.8 percent of niccolite, in contrast to the inclusion type of sulfide grains which have 17.6 and 10.8 percent of platinum-group minerals and 22.0 and 10.8 percent of niccolite. Also, the sulfide-phase assemblages are more complex and abundant within the postcumulus materials than they are within inclusions.

COMPARISON OF TYPES OF SULFIDE OCCURRENCE

The principal similarity between the sulfide occurrences in the G and H zones is the dominance of pentlandite, pyrrhotite, and chalcopyrite in both zones. Less than 10 percent of the sulfide grains and aggregates have neither pentlandite nor pyrrhotite present. Table 3 gives the sulfide-phase abundance by assemblages for the G and H zones. Besides the outstanding differences between the four textural categories in which the sulfides are associated with cumulus, postcumulus, or alteration minerals, there are differences in the amounts present in each category and in their orientation, grain size, sorting coefficients, and mutual

TABLE 3.—Sulfide abundances in the G and H chromitite zones

[Mineral symbols: pn, pentlandite; po, pyrrhotite; nc, niccolite; Pt, platinum-group minerals; ccp, chalcopyrite; mo, molybdenite; hz, heazlewoodite; vl, violarite; il, ilmenite; mt, magnetite]

Mineral assemblage	Abundance (percent)	
	G chromitite zone	H chromitite zone
pn	54.2	53.5
po	25.0	25.1
ccp	2.7	.8
vl	---	.1
nc	2.7	1.5
Pt	3.3	1.3
mo	---	.1
hz	.4	---
pn+po	3.7	8.8
pn+hz	---	.6
pn+phase Q	.3	1.0
pn+ccp	.4	.6
pn+nc	.5	.3
po+vl	---	.1
po+ccp	2.2	.7
po+Pt	---	.1
po+mt	.1	1.1
po+hz	.3	---
ccp+mt	.4	.2
pn+il	---	.1
nc+mt	---	.1
pn+mt	.7	.3
hz+mt	.1	---
pn+po+phase Q	---	.5
pn+po+ccp	1.0	1.3
pn+hz+Pt	---	.1
po+ccp+Pt	---	.1
pn+po+Pt	.1	.1
pn+po+mt	1.1	.5
pn+phase Q+mt	---	.1
pn+Pt+mt	---	.1
pn+hz+mt	.1	---
pn+po+ccp+mt	.4	---
pn+po+phase Q+ccp	.1	---
po+phase R+phase S	---	.2
pn+po+phase Q+Pt	---	.1
pn+po+phase Q+mt	---	.3
pn+po+ccp+Pt	---	.1
pn+po+nc+mt	---	.1
pn+po+ccp+Pt+mt	---	.1
pn+po+ccp+vl	.1	---

phase boundaries in multiphase grains. The nearly universal presence of pyrrhotite and pentlandite in the four categories and two zones suggests a similar ultimate source for the sulfide minerals, whereas the differences imply different mechanisms for the localization of the phases for each category.

Although there are broad similarities in the phases present as inclusions and associated with postcumulus minerals, the platinum-group minerals and niccolite rarely occur as polymineralic grains and are concentrated as inclusions in cumulus phases, whereas the rare platinum-group minerals in sulfides associated with postcumulus minerals generally form composite grains. Inclusions also contain fewer polymineralic grains than are present in the sulfides associated with the postcumulus minerals. No four- or five-phase grains are present as inclusions. Spongy pyrrhotite

and pyrrhotite forming wormy intergrowths in pentlandite have not been observed as inclusions but they form an important part of the sulfide minerals occurring with postcumulus phases.

The study of the orientation of grains relative to a stratigraphic top or bottom shows that sulfides in inclusions tend to have a greater degree of preferred orientation with respect to layering than those that occur as interstitial material. The sulfide grains in inclusions, on the average, are four to five times smaller than the interstitial sulfide grains and the sulfide

grains in inclusions show better sorting characteristics than sulfide grains in the other textural categories.

The relatively complex phase assemblages and intricate contacts between sulfide minerals in multiphase grains that occur associated with postcumulus minerals are in sharp contrast to the lack of these characteristics for the inclusion subcategories. This fact implies that if gravitative settling is important in their origin, then reactions and changes have occurred since they accumulated in the interstices.

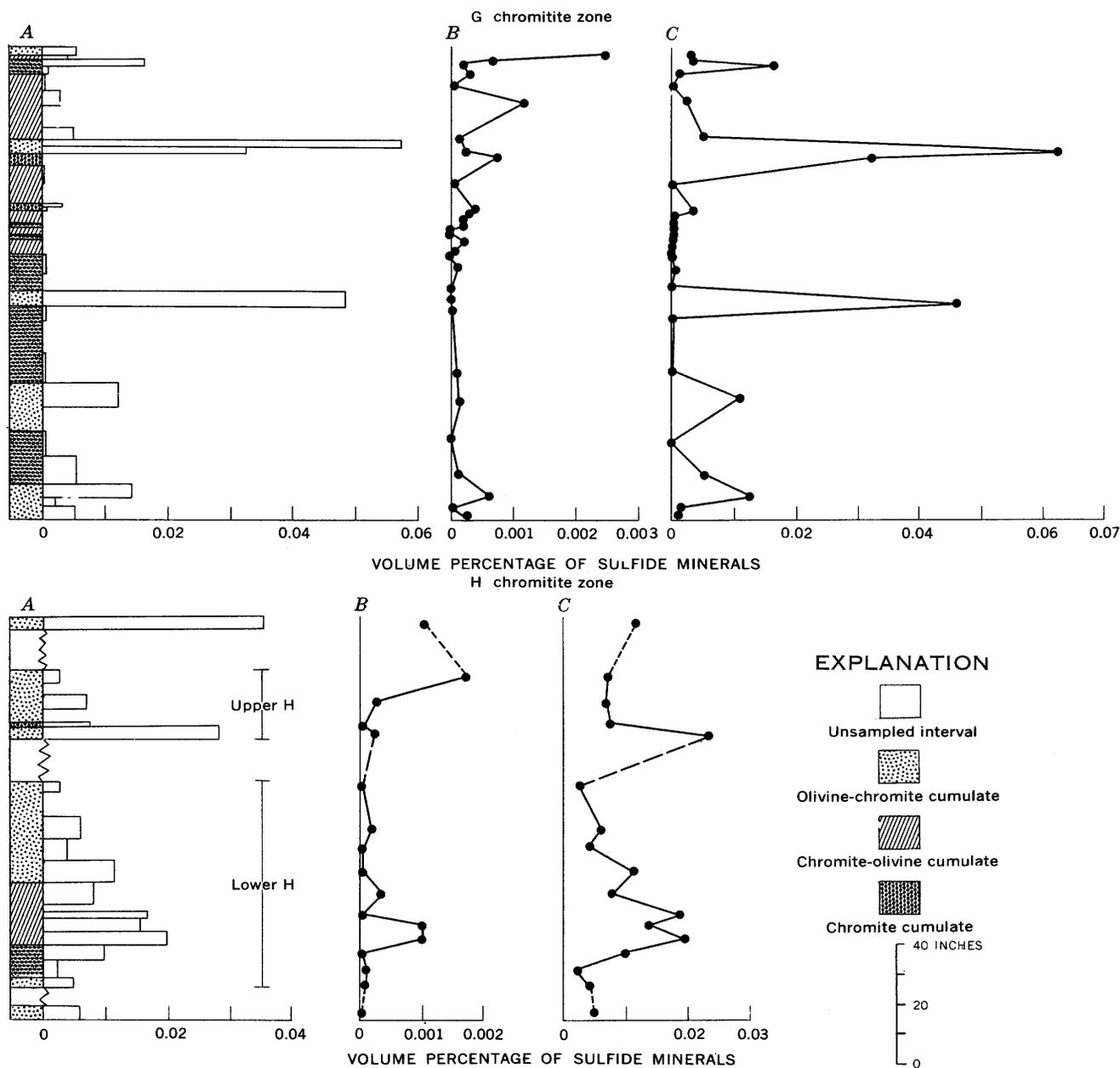


FIGURE 6.—Variation of volume percentage of sulfide minerals correlated with stratigraphic position. A, Total sulfide minerals. B, Sulfide inclusions in cumulus minerals. C, Sulfide minerals associated with postcumulus minerals.

VARIATION OF SULFIDE CATEGORIES WITH STRATIGRAPHY

Figure 6 presents the variation in volume percentage of total sulfide minerals (A) for the G and H zones plotted against stratigraphic position. Although more sulfide minerals are concentrated in the chromitite zones than elsewhere in the Ultramafic zone, they are not particularly numerous in the cumulus chromite layers of these zones, at least in the localities where the samples were taken. Instead, layers of olivine-chromite and chromite-olivine cumulate tend to have relatively higher amounts of sulfide minerals.

The overall pattern of maximum sulfide content for both zones is similar, neglecting samples taken from the H zone below the footwall and above the hanging wall. The pattern consists of a general paucity of sulfide minerals at the bases of the zones, an increase to a maximum near the central parts of the zones, and a decrease to a minimum in the hanging walls. Here the similarity between the two zones ends; the variability within the G zone is much larger than the variability within the H zone because some layers contain relatively large amounts of sulfide minerals and some none. Little gradation between sulfide-rich layers and sulfide-poor layers in the G zone is observed. However, in the H zone, each maximum concentration is formed by a gradation from sulfide-poor layers to sulfide-rich layers. What the sulfide distribution relations are between the upper and lower parts of the H zone is unknown. The lower part of the H zone begins with a low sulfide concentration which increases gradually to a maximum concentration and then decreases in the direction of the hanging wall. The base of the upper part of the H zone has a high sulfide concentration which diminishes toward the hanging wall.

In figure 6, the volume percentages of sulfide inclusions in cumulus mineral (B) and sulfide minerals associated with postcumulus minerals (C) are plotted against stratigraphy for the G and H zones. The broad distribution patterns are similar to total sulfide mineral volume percentages, but in detail many differences exist.

The volume percentages of sulfide inclusions in cumulus minerals (fig. 6B) are similarly distributed in the stratigraphic sequence in both the G and H zones. In general, sulfide inclusions are fairly abundant near the base of each zone, are rather constant in amount through the middle of the zone, and increase to a maximum volume of inclusions at the hanging walls of each zone. Sulfide inclusions in olivine constitute the largest volume percent of sulfide inclusions in the upper part of each zone.

Sulfide minerals that are associated with postcumulus minerals (fig. 6C) account for the majority of the sulfide grains present; therefore, their stratigraphic distribution pattern, in volume percent, corresponds closely with the total volume percent of all sulfide minerals within both zones. If this textural type of sulfide grain were plotted by subcategories, similar distribution patterns would be obtained.

When the average diameter of all sulfide grains within an individual sample is plotted versus its stratigraphic position (fig. 7), a pattern remarkably similar to that of the total sulfides (fig. 6A) is obtained. This correlation is more striking in the H zone than in the G zone where large numbers of micron-size grains affect the weighted average of the diameters. The average diameter of sulfide inclusions oscillates between large and small within both zones, but the trend is from small-sized inclusions at the bases of the zones to large-sized inclusions at the tops of the zones. However, the average diameter of sulfide grains and aggregates associated with postcumulus phases is largest where the volume percentage of sulfide grains is large. Regression analyses of the relation of grain size of a particular textural category of sulfide versus the volume percent present confirm the general impression of correlation between grain size of sulfides and volume percent. Correlation coefficients ranged from 0.80 to 0.95 at levels of significance greater than 99 percent.

If the variations in percentage of the various minerals present are examined from the bases of the zones to their tops, then the patterns are those shown in figure 8. Inclusion-type sulfide minerals and sulfide minerals associated with postcumulus silicate material have been plotted separately. Abundance data on sulfide volumes present for the G zone (figs. 8A, C) tend to show more scatter and less regularity than those for the H zone (fig. 8B, D). This probably results from lesser statistical significance of fewer sulfide grains.

More inclusions in the lower part of the H zone (fig. 8B) contain pentlandite than in the upper part. The trends of pentlandite concentration in the G zone (fig. 8A) are opposite—pentlandite increases in abundance upward across the zone. Pyrrhotite gradually decreases in abundance in the upper parts of both zones. Platinum-group minerals and niccolite appear to be more irregular in abundance than pyrrhotite and pentlandite. In the H zone, niccolite increases up section as platinum-group minerals decrease; in the G zone, platinum minerals and niccolite are present in the lower part of the section, increase upward to a maximum, then gradually decrease.

The abundance of pentlandite and pyrrhotite associated with postcumulus phases (fig. 8C, D) is simi-

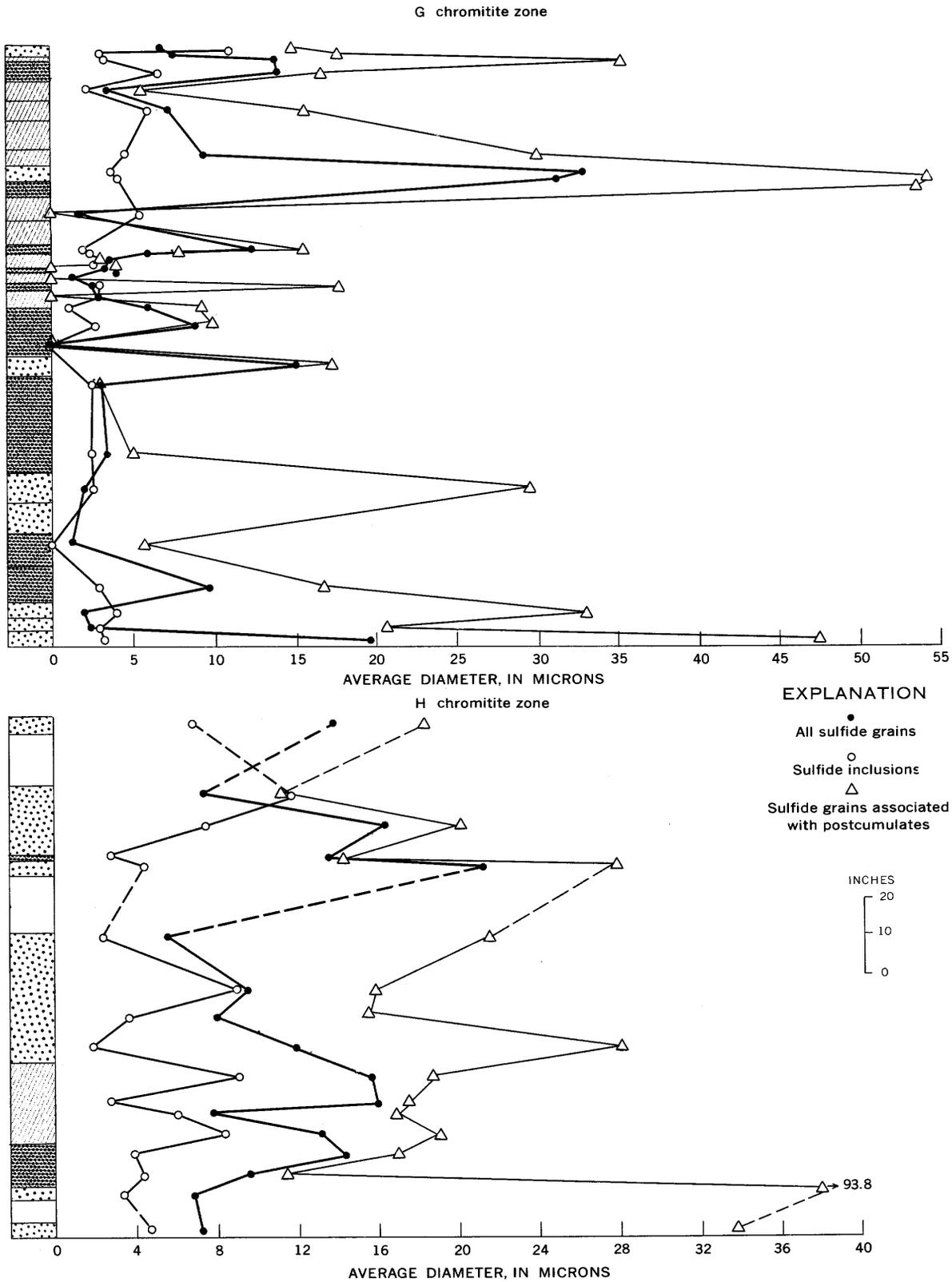


FIGURE 7.—The size (average diameter) of sulfide grains and aggregates correlated with stratigraphic position. Symbols on the stratigraphic section are the same as in figure 6.

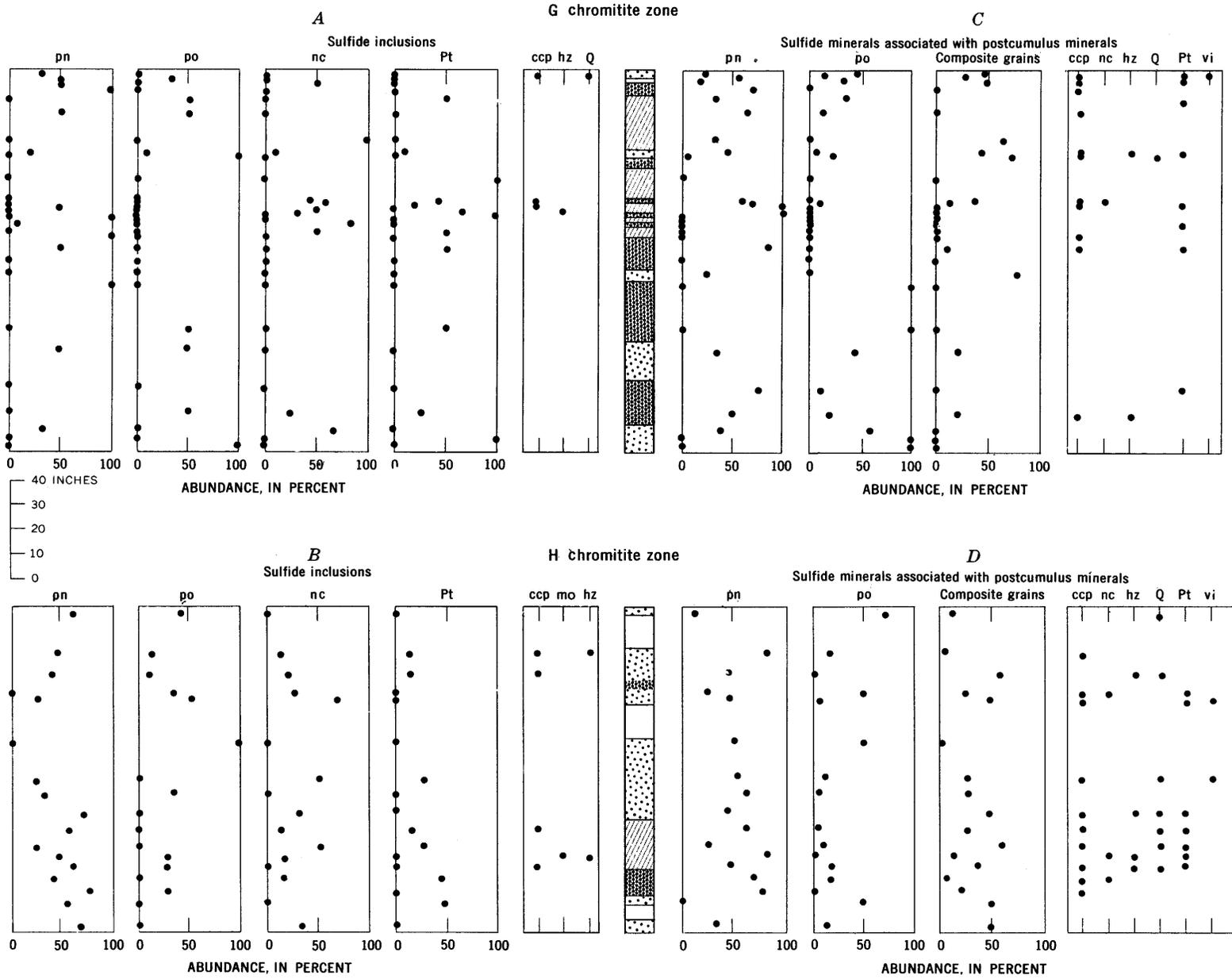


FIGURE 8.—Variation of abundance of phases correlated with stratigraphic position. *A*, Sulfide inclusions, G zone. *B*, Sulfide inclusions, H zone. *C*, Sulfide minerals associated with postcumulus minerals, G zone. *D*, Sulfide minerals associated with postcumulus minerals, H zone. Symbols in the stratigraphic section are the same as in figure 6.

lar to the distribution of the total volume percent of sulfide minerals within the zones. Pentlandite abundance varies inversely with that of pyrrhotite.

GENETIC IMPLICATIONS

One hypothesis for the origin of the sulfide minerals in the chromitite zones is that the minerals were produced from the same magma that produced the silicates; the sulfur either came from sulfur dissolved in the magma, in immiscible sulfide liquids, or in crystalline solid solutions. This hypothesis has been offered to explain sulfide occurrences within the Bushveld Complex (Wagner, 1924), the Insizwa intrusive (Scholtz, 1936), the Skaergaard intrusion (Wager and others, 1957), the Sudbury intrusive body (Hawley, 1962; Naldrett and Kullerud, 1967), the Muskox intrusion (Chamberlain, 1967), and many other occurrences including the modern Hawaiian lavas (Desborough and others, 1968). If this general hypothesis is correct, then the observations on the sulfide mineralogy of the G and H zones corroborate it and should also contribute to an understanding of how the process works. Before examining the observations in detail, a few general definitions of concepts and terms are needed.

Three stages in sulfide mineral formation should be distinguished: (1) the separation and concentration of sulfide material from the basaltic magma, (2) subsolidus reactions of the sulfide material within itself or with oxide or silicate phases, and (3) later redistribution of sulfide phases by alteration processes such as serpentinization. Only stages 1 and 2 are of importance in this paper. The actual phases observed in the G and H chromitite zones are dominantly a result of stage 2 processes.

Three magmas are described. One is the basaltic magma which forms the reservoir from which the cumulus minerals of the cyclic units crystallized. Within the basaltic magma there is the possibility of a second magma, an immiscible sulfide-metal-oxygen liquid, which will be referred to as sulfide liquid or immiscible sulfide droplets. The third magma is of the same basaltic composition as the reservoir, but this magma was trapped in the pore spaces between the cumulus phases and soon was separated from the reservoir. It will be called trapped magma. It solidified to form part of the postcumulus material.

The basaltic magma that formed the cyclic units of the G and H chromitite zones was probably introduced into its position above the previous cyclic units by a mechanism of variable-depth convection and periodic refreshment of the bottom magma (Jackson, 1961, p. 96-99). The temperature of this magma was at or

above the crystallization temperature of olivine and chromite. Olivine and chromite pairs indicate temperatures during crystallization of perhaps as much as $1,425^{\circ} \pm 300^{\circ}\text{C}$ near the base of the chromitite zones and as low as $903^{\circ}\text{--}1,100^{\circ}\text{C}$ near the top of the chromitite zones (Jackson, 1969). The cumulus minerals chromite and olivine and their sulfide mineral inclusions record the liquidus history of crystallization of the basaltic magma of the reservoir and its subsolidus history. Sulfide phases associated with postcumulus silicate phases may reflect the liquidus and subsolidus history of both the reservoir basaltic magma and the trapped basaltic magma as well as the interaction between the two.

The intimate association of sulfide minerals within cumulus phases and with postcumulus phases in the Stillwater Complex supports the thesis that sulfide material was present at the time and was under the conditions of silicate crystallization from the basaltic magma. The introduction of small sulfide mineral blebs into the centers of cumulus phases at any other time than that of crystallization is difficult to accept. Furthermore, the presence of different stratigraphically controlled sulfide populations inside cumulus minerals and their repetition in at least two zones leads me to believe that the sulfide minerals are directly related to the separate reservoir magmas and, in fact, were derived from the magmas.

Assuming that the sulfide, oxide and silicate minerals in the chromitite zones are comagmatic, the sulfur must have been dissolved in the magma, in an immiscible sulfide liquid, in crystalline solid solutions, or held in a combination of these. The contribution of these sources must be evaluated.

The average volume percent of sulfide minerals in chromitites is on the order of 0.01. This concentration in the magma is compatible with the solubility of 0.1 weight percent of sulfur in a thoeilitic basalt from the Alae lava lake (Skinner and Peck, 1969) and 0.06 weight percent of sulfur estimated for the Skaergaard magma (Wager and others, 1957). MacLean (1968) has shown experimentally that 3-5 weight percent of sulfur can be dissolved at very low oxygen fugacities in iron silicate melts.

Although this appears to be a sufficient source for sulfur, observations from the chromitite zones suggest extreme heterogeneity in the sulfur content of the magma. First, the proportions of sulfide to silicate minerals within individual inclusions in cumulus olivine and chromite range from 0 to 100 percent. If the inclusions were trapped by olivine and chromite at the basaltic magma liquidus, then the basaltic magma must have contained immiscible sulfide droplets or

sulfide crystals. Second, the amount of trapped magma varies from about 4–43 percent by volume in the chromitite zones and shows no correlation with the volume of sulfide minerals associated with the postcumulus minerals. Third, local massive sulfide lenses occur conformable to the base of chromitite zones, indicating a larger amount of sulfur in the basaltic reservoir magma than the average volume percent of sulfide minerals in the chromitite zones would suggest.

Either immiscible sulfide liquids or crystalline sulfide solid solutions can account for the apparent inhomogeneity in the reservoir basaltic magma as reflected by the observed sulfide mineralogy. Sulfur dissolved in the reservoir magma cannot alone account for the inhomogeneity. If immiscible droplets were the only source of the single-phase and multiple-phase grains with varying proportions of different minerals, many different compositions of sulfide liquids would be required. Because the distribution of metals in the sulfide liquid was most certainly controlled by the reservoir basaltic magma, it is impossible to accept the theory of many different sulfide liquids. Jackson's (1969) liquidus temperatures of 1,400°C in the lower portions of the chromite zones, based on olivine-chromite pairs, are too high to permit crystalline phases to be present if compared with the 1,100°–1,200°C temperatures at which crystalline nickel-rich pyrrhotite solid solutions are present in experimental studies (Craig and Kullerud, 1969; Kullerud and others, 1969). Because immiscible sulfide droplets alone cannot account for the observed sulfide mineral heterogeneity, I am led to conclude that soon after olivine and chromite began to crystallize from the magma, significant amounts of sulfur must have existed in both crystalline solid solutions and immiscible droplets and that Jackson's estimates of liquidus temperatures are too high. This conclusion is contrary to the inferences which may be drawn from Jackson's temperatures.

In the above discussion, it is implied that the reservoir basaltic magma is the main source of the sulfide minerals. Evidence for this is that the amount of postcumulus silicates (representing trapped magma) varies from 4 to 43 percent by volume and shows no correlation with the amount of sulfide minerals associated with postcumulus material; hence, contributions from the trapped magma were small. The composition of the trapped magma could be affected by the reservoir magma as long as there was permeability between the two magmas in the crystal pile. The amount of permeability was probably small; the distance over which the crystal pile was permeable was on the order of feet or inches, as evidenced from soft, sedimentary, deformation features (Hess, 1960; Jackson, 1961).

As the pile cooled and crystallized, the trapped magma could generate more sulfide liquid, which then could react or add to the preexisting sulfide concentrations in the pores of the olivine-chromite cumulates. This magma, after cooling below the temperature of crystallization of a nickel-rich pyrrhotite solid solution, could yield sulfides as crystalline material. The soluble sulfur in the magma must have eventually reacted with the concentrations of sulfides already present, but considering the low solubilities of sulfur in basaltic magmas and the amount of trapped magma, any contribution to them must have been small. If this is what happened, it means that the major processes took place in the reservoir magma.

If the above model is valid, then observations on the sulfide minerals suggest several possible embellishments. Single phase inclusions of laurite, platinum-group sulfide minerals and alloys, niccolite, and molybdenite suggest that these were trapped by the cumulus minerals after the inclusions existed as crystals. Multiple-phase inclusions may have been trapped as Fe-Ni-Cu-S crystalline solid solutions or as immiscible droplets which, upon cooling, crystallized and reacted to form the observed assemblages. After any of these appeared in the magma, they must have grown in size, because the average diameter of the inclusions increases from the base to the top of the chromitite zones.

This hypothesis cannot account for the single-phase grains of pentlandite and pyrrhotite found as inclusions in cumulus phases. Experimental studies indicate that pentlandite does not form from a liquid; only Fe-Ni monosulfide solid solutions do (Kullerud and others, 1969). If such a solid solution were the phase trapped by the cumulus minerals, then it is possible, but not proven, that an exchange of Ni or Fe between host and inclusion could form pentlandite or pyrrhotite. Assuming that a 10-micron pentlandite inclusion with a density of 4.91 gm per cm³ was derived from an inclusion of Fe_{0.5}Ni_{0.5}S, only 9.7×10⁻¹¹ grams of Ni and 9.3×10⁻¹¹ grams of Fe would have to be added to form pentlandite with a composition of Fe_{4.5}Ni_{4.5}S₈. Unpublished analyses of the cumulus phases (E. D. Jackson, oral commun., 1968) show that there is enough Ni and Fe to do this. Because of the small amounts of material involved, any zoning that might have been produced in the host by this mechanism remains undetected. If this process is not accepted, then the present phase diagrams are not applicable to a differentiating magma, and one must assume that a dense phase of pentlandite can exist at magmatic temperatures. All evidence from studies of Hawaiian lava agrees with the experimental studies.

More abundant sulfide grains in phases associated with postcumulus material must have had at least two sources—cumulates from the reservoir basaltic magma and minor postcumulus growth from the trapped magma. Determination of the source of each sulfide grain is impossible, but the three subcategories of sulfide grains associated with postcumulus material suggest at least two sources. Also, the poor sorting exhibited by grains associated with postcumulus material could be interpreted as evidence for multiple origins for the grains. Sulfide grains that completely fill the pore spaces and that are molded around the cumulus phases (subcategory 1) possibly accumulated as immiscible droplets or crystalline solid solutions contemporaneously with olivine and chromite. The sulfide grains that have sharp contacts with silicates could be interpreted as cumulates because they are properly oriented in the pores and have local growths of postcumulus chromite or olivine around them (subcategory 2), but they could also represent accumulated droplets. Orientation studies of grains in the G and H zones show that grains with these characteristics could amount to 60 percent of subcategory 2. The rest of this subcategory and possibly the third subcategory of ragged grains appear to represent sulfide droplets and crystalline phases that were formed by the "trapped magma" as the pile cooled. Grains with ragged edges might result from addition of material from the last sulfur dissolved in the magma or from reactions that may have taken place. By abundance, about 84 and 64 percent of the sulfide grains of the G and H zones respectively fall in subcategories 1 and 2.

The number of sulfide grains representing all textural categories correlates closely with the volume of sulfide minerals present in a particular horizon. (Note that this relation is not necessarily true; there could be one grain of 10-micron diameter present in one rock and 1,000 grains of 1-micron diameter in the other rock, but both would contain the same volume percent of grains.) The well-sorted nature of the inclusions and their size variations suggest that they were distributed in the magma chamber by some sort of crystal-settling phenomena. For these reasons, sulfide distribution must have the same relationship to the rate of production and growth of these inclusions as it has to the rates of accumulation and growth of cumulus olivine and chromite. Jackson (1961) suggests that olivine and chromite accumulated on the bottom of the magma chamber as rapidly as they were able to grow and nucleate. Settling was rapid. Reexamination, using viscosities given by Shaw, Wright, Peck, and Okamura (1968) for a basaltic magma, shows that olivine and chromite crystals of the size range found

in the G and H zones² would settle at rates of thousands to ten thousands of centimeters per year, whereas inclusions with a 5-micron radius would settle at rates of tenths to hundredths of a centimeter per year. The change in size of inclusions as a function of stratigraphic position must reflect the growth and nucleation rates of sulfide grains and interrelate with the amount of time required for them to be concentrated at a particular horizon by settling.

The number of sulfide grains associated with postcumulus phases also correlates well with the amount of sulfides. Aggregates and grains with 20-micron radii would settle at rates of between 1/10 and 1 centimeter per year. Comparison of these values with rates of settling for olivine and chromite implies that nucleation and growth, rather than settling, were also the major controls of their concentration. This theory assumes that the grains settled as entities and were not attached to chromite and olivine or caught up in currents or eddies controlled by the settling olivine and chromite. Observations of the orientation of sulfide grains suggest that the sulfide grains may have been attached to olivine and chromite, but not enough evidence is available to evaluate this feature.

If, instead of the smooth variations in temperature and partial pressure of oxygen initially suggested by Jackson (1969), the actual variations in temperature and pressure fluctuated, as indicated by Jackson's (1969) detailed data, then it might be possible to explain the distribution of sulfide minerals in another manner. The relation between the sulfide volume percentage, the $Fe^{+3}/(Fe^{+2} + Fe^{+3})$ ratio in chromite, and the temperature of the liquidus from olivine and chromite pairs is shown in figure 9. There is a suggestion from these data that higher temperatures at intermediate values of oxidation ratios favor the accumulation of sulfide minerals. Plots of temperature against oxidation ratio, contoured with volume percent of sulfides, suggest that this correlation is not strong. Smith (1961) suggested that a low oxidation potential in a mafic magma would favor the formation of sulfide liquids and that higher temperatures would favor sulfur dissolved in the magma. MacLean's (1968) experimental work indicates that the solubility of iron sulfide in an iron silicate melt is at a maximum when coexisting with iron and that solubility decreases with increasing oxygen fugacity. Evaluation of the Stillwater sulfide occurrences neither supports nor refutes this evidence, but it does indicate that in order to unravel temperature and oxidation effects, effects of

² Assumed values: sulfide density, 4.90 g per cm³; olivine density, 3.30 g per cm³; chromite density, 4.55 g per cm³; magma density, 2.65 g per cm³; and magma viscosities, 500 and 8,000 poises.

growth, settling, and nucleation rates and the various sources of sulfur must first be understood.

Hess (1960) gives fairly constant values of 0.04 weight percent of sulfur in the various zones of the Stillwater Complex. The Skaergaard intrusion (Wager and others, 1957) and the Muscox intrusion (Chamberlain, 1967) also show a constant or slightly increasing total sulfur content with higher stratigraphic position. A chromitite zone is a small segment of a complex and, moreover, is not rich in sulfur; therefore, it is suspected that the amount of sulfur in the magma remained nearly constant during the deposition of a chromitite zone.

However, the ratios of phases have been shown to change within the horizons. Because pyrrhotite and pentlandite are the most abundant sulfide minerals present, the most significant variation to be considered is the distribution of nickel and iron in sulfides from the bottom to the top of a zone. The average pentlandite to pyrrhotite-plus-pentlandite ratio for both the G and H zones is about 0.68, so that bulk composi-

tions and the process of differentiation which yielded the sulfides were similar in both zones. The lower H zone may be used as an example for interpretation although the data may not be representative of the whole differentiation and accumulation process. The G zone offers less data of statistical significance in the distribution of sulfides and the data seem to be more variable, not only with respect to the sulfide amounts, but also with respect to compositions of chromite and olivine (Jackson, 1969).

Nickel and iron have complementary distributions in the sulfide phases; therefore, only nickel will be considered here. The data suggest, in general: (1) the abundance of pentlandite decreases from the base to the top of the lower H zone, (2) the abundance of pentlandite in single-phase grains decreases upwards and the number of single-phase grains decreases upwards, and (3) the abundance of pentlandite in multiphase grains decreases upwards but the number of these multiphase grains increases upwards. This evidence implies that the weight percent of nickel in all

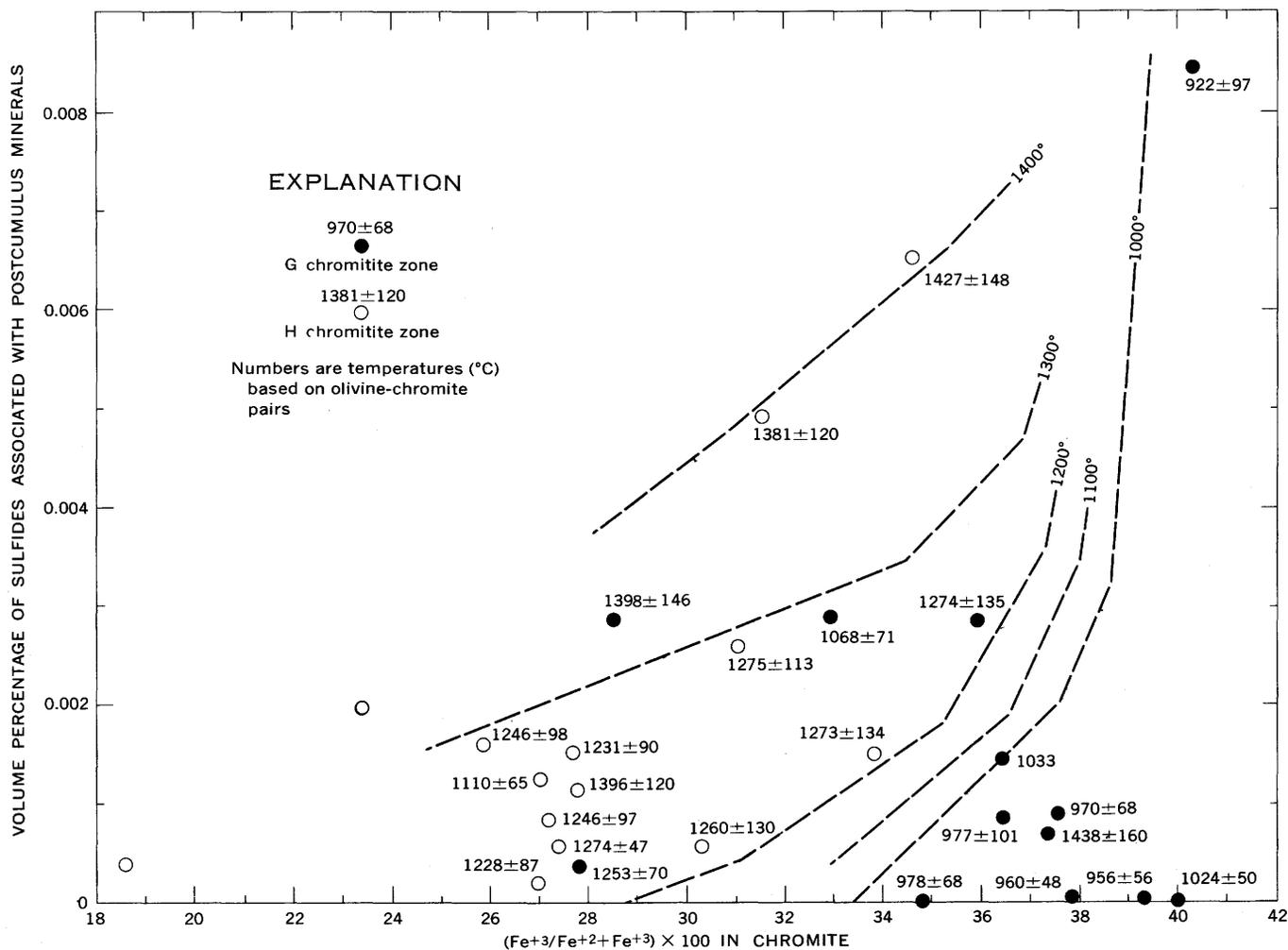


FIGURE 9.—Volume percentage of sulfide minerals versus Fe³⁺/(Fe²⁺+Fe³⁺) ratio in chromite.

sulfide grains decreases from the base to the top of the lower H zone and suggests a nickel impoverishment in the source liquids.

CONCLUSIONS

The sulfide grains found in the G and H chromitite zones came from the same magmas that produced the respective cyclic units of the zones. Sulfide minerals occur as sulfide inclusions in cumulus chromite and olivine and as sulfide phases in interstices between cumulus phases. Studies of texture, size, phases, abundance, and orientation indicate that sulfides existed in the magma batches in three forms: as crystalline phases suspended in the basaltic magma; as immiscible sulfide liquids; and as sulfide soluble in the basaltic magma. During differentiation, nickel and sulfur were preferentially concentrated in the sulfide crystals and liquids; the nickel depletion of the magma with time is shown by the sulfide phases. Platinum-group metals were preferentially concentrated in cumulus chromite as inclusions of sulfides and alloys, and at the time of entrapment they probably existed as crystalline phases.

Although temperature and partial pressures of oxygen and sulfur were the main controls for the formation of sulfide phases, nucleation, growth, and settling rates probably determined the present distribution and concentrations of sulfide minerals in the G and H chromitite zones.

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