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STRATIGRAPHIC AND LATERAL VARIATION OF CHROMITE COMPOSITION IN THE STILLWATER COMPLEX[1]

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ABSTRACT

Layered concentrations of chromite occur at 13 recognizable stratigraphic horizons within the 3500-foot thick Ultramafic zone of the Stillwater Complex. Five of the chromitite layers or zones can be recognized for 15 miles along strike, and one can be traced nearly 30 miles. Samples of chromitite from the footwalls of these zones were collected at intervals of about 2.5 miles along strike and 43 chemical analyses of clean chromite were made.

Considered individually, the major cations Cr^{3+}, Al^{3+}, Fe^{3+}, Mg^{3+}, and Fe^{2+} in the chromite show no simple vertical or lateral patterns of variation. However, total iron ($Fe^{2+}+Fe^{3+}$) in the analyzed samples decreases regularly from about 6.0 cations/unit cell in the lower zones to about 4.5 cations/unit cell in the middle zones, then changes, and increases regularly to about 5.5 cations/unit cell in the upper zones. Lateral variations in the total iron content are small, so that isochemical lines trend parallel to the layering. The oxidation ratio ($Fe^{3+}/Fe^{2+}+Fe^{3+}$) of the chromite, on the other hand, shows a strong lateral variation, decreasing from about .30 at the eastern and about .15 at the western ends to less than .05 in the central part of the complex. Vertical variation of the oxidation ratio in any one stratigraphic section is small and irregular, so that isochemical lines trend at steep angles to the layering. Inasmuch as total numbers of trivalent and bivalent cations per unit formula of balanced chromite are fixed, the combination of constant total iron and changing oxidation ratio of chromite along any given chromitite zone accounts for the complex variation of the individual cations.

The strong lateral change in oxidation ratio of the chromite suggests that a lateral oxygen partial pressure gradient persisted in the magma during crystallization and accumulation of the Ultramafic zone. The early decrease in total iron between the chromitite zones suggests an early reversal in the expected trend of differentiation, followed in the upper part of the stratigraphic section by normal iron-enrichment.

PURPOSE AND SCOPE

The compositions of the primary precipitate minerals of the Ultramafic zone of the Stillwater Complex are being studied for the purpose of describing and interpreting the crystallization history of the intrusion. Three primary precipitate minerals occur in the Ultramafic zone: olivine, bronzite and chromite. Of these, chromite is the least abundant but most interesting because it is the only one capable of containing major amounts of trivalent cations. This paper presents some preliminary results of a study of the lateral and vertical changes in chromite composition within the Ultramafic zone of the complex.

GENERAL SETTING OF THE CHROMITE

The Stillwater Complex can be divided into several major stratiform zones (Jones *et al.*, 1960, p. 286–289; Hess, 1960, p. 49–50; Jackson, 1961, p. 2–4). The Ultramafic zone, which lies in the lower part of the complex and has an average stratigraphic thickness of about 3500 feet, can be subdivided into a Peridotite member and a Bronzitite member (Fig. 1). Virtually all the chromite in the complex occurs within the Peridotite member of the Ultramafic zone.

On the basis of mapping at scales of 1:12,000 and 1:1200 the Peridotite member has been divded into 15 subunits, called cyclic units, characterized by layers of harzburgite, chromitite, and bronzitite arranged in a particular stratigraphic order (Jackson, 1961, p. 13–15). A typical cyclic unit is shown in Fig. 2. The sequence of rock types is poikilitic harzburgite, chromitite, olivine chromitite, poikilitic harzburgite, granular harzburgite and bronzitite. In terms of the primary precipitate (or settled minerals) the sequence has been interpreted as recording the continuous crystallization and deposition first of olivine, then chromite, olivine plus chromite, olivine, olivine plus bronzite and finally of bronzite alone. The sharp breaks between cyclic units are thought to represent interruptions of crystallization and deposition, followed by a repetition of the cycle to form the succeeding unit. Each of the cyclic units is distinctive in thickness, proportion of rock types, grain size or textural peculiarities.

DISTRIBUTION AND CORRELATION OF THE CHROMITITE ZONES

Thirteen of the cyclic units contain recognizable layered concentrations of chromite. These concentrations, locally called zones, are compound; they are usually composed of massive chromitite layers at the base, giving way to alternating olivine chromitite layers and massive chromitite layers upward

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[1] Publication authorized by the Director, U. S. Geological Survey.

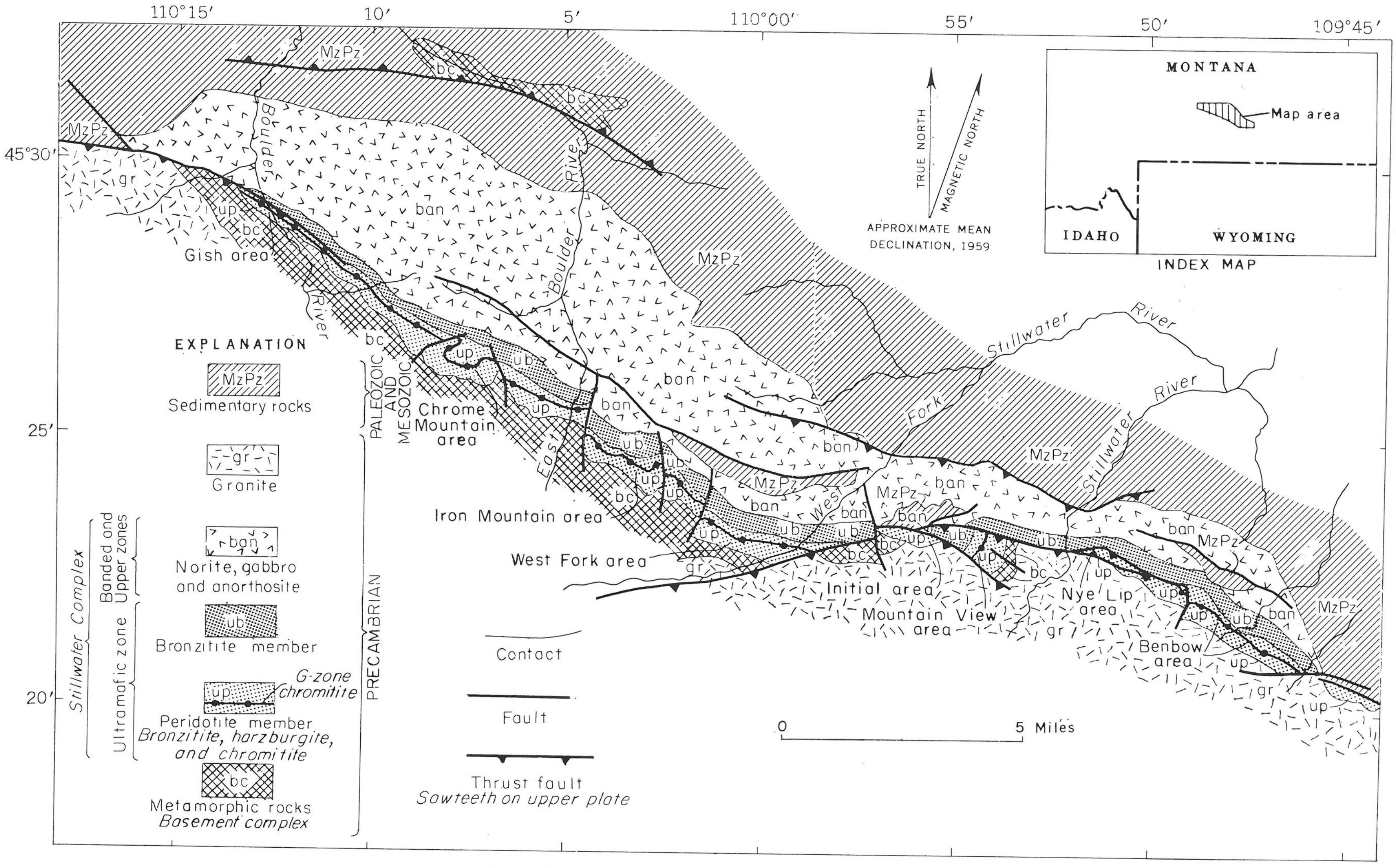

FIG. 1. Geologic index map of the Stillwater Complex.

in the section. The chromitite zones have been mapped, correlated by field criteria, and lettered more or less sequentially from the base upward. A correlation diagram of the chromitite zones of the Peridotite member is shown as Fig. 3. Several basins in the Peridotite member can be recognized on the correlation diagram, although they are exaggerated by the expanded vertical scale. Both cyclic units and chromitite zones are thicker in the basin sections than on the shelf sections at either side.

Five of the chromitite zones, the A, B, G, H and K, can be traced for 15 miles along strike, and one, the G, can be traced nearly 30 miles. The G is also the thickest chromitite zone, ranging from a maximum of about 12 feet to a minimum of about 1½ feet in thickness. The H zone is generally about half as thick as the G, and is followed in decreasing order of thickness by the B, A and K zones. The remaining zones are generally only one or two inches thick, but even so, several of them (for example, the I and J zones) can be traced more than ten miles.

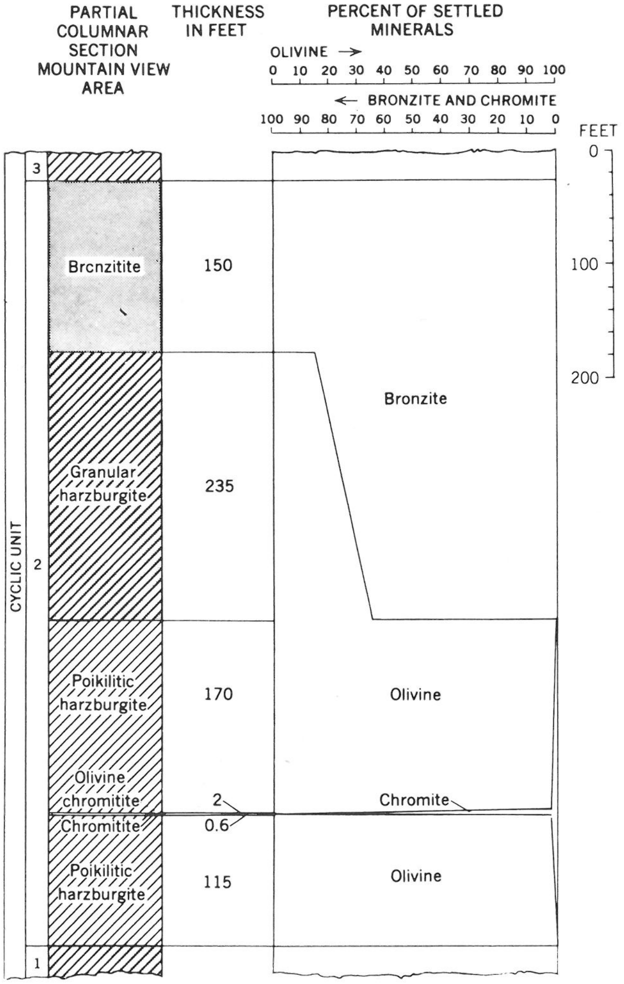

FIG. 2. Stratigraphic variation of settled minerals in a typical cyclic unit of the Peridotite member.

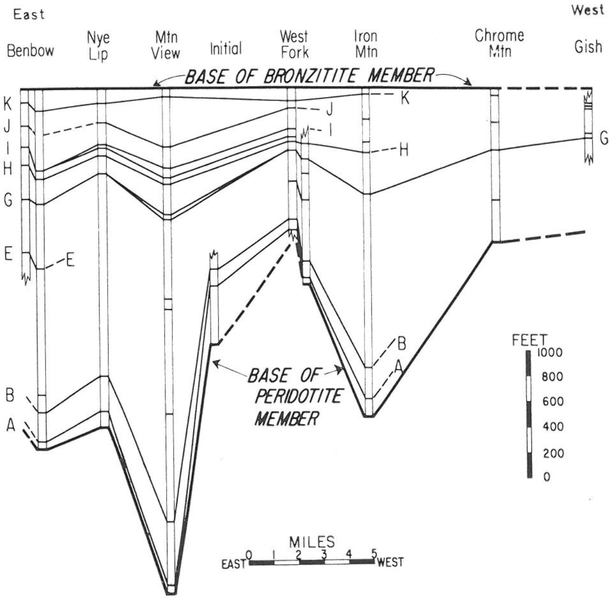

FIG. 3. Correlation of chromitite zones in the peridotite member, Stillwater Complex.

SAMPLING AND ANALYSIS OF THE CHROMITE

Ninety-five chromite-bearing samples were collected from the layered chromitites of the complex to determine changes in composition of chromite in three ways: vertically within single zones; stratigraphically between zones; and laterally along zones. Forty of the samples were selected to determine the within-zone variations, 43 to determine the between-zone and lateral variations, and the remainder for various special purposes.

The samples taken to investigate between-zone and lateral variations were collected from 8 areas (Fig. 1) at intervals of about 2.5 miles along the strike of the intrusion. Samples were collected from the footwalls of all the exposed chromitite zones in each area (Fig. 3).

Chromite samples were cleaned of silicate minerals, examined in polished grain mounts to establish purity, and chemically analyzed by a method developed by Dinnin (1959). Minor amounts of impurities were subtracted from the analytical values and the analyses were recalculated to cations per unit cell on the assumption of balanced charge. None of the analyzed chromites showed a cation deficiency or excess.

RANGE OF CHEMICAL COMPOSITION OF THE CHROMITE

Five major cations—Mg^{2+}, Fe^{2+}, Cr^{3+}, Al^{3+} and Fe^{3+}—comprise more than 98 per cent of the total cations in the analyzed chromite samples, so the major variation can be expressed graphically on a triangular prism (Thayer, 1956, p. 16). Fig. 4 shows the range in the 95 samples of variation of the five major constituents expressed as end-member spinels on such a prism. The field of occurrence forms a cylinder whose axis extends from the Cr-Al face of the prism toward the magnetite corner. The prism also illustrates the interdependence of the major cations in the chromite. The sum of Mg^{2+} and Fe^{2+} is virtually a constant, so that as one increases, the other must automatically fall. Similarly, the sum of Cr^{3+}, Al^{3+} and Fe^{3+} is nearly constant, so that an increase in one will generally result in a decrease in the other two. However, the charge balance does not require such a relation between cations of the R^{2+} and R^{3+} groups, and interactions between individual cations such as Fe^{2+} and Fe^{3+} may vary independently.

VARIATIONS IN COMPOSITION OF THE CHROMITE

The variation in composition of chromite within single zones has been reported elsewhere (Jackson, *et al.*, 1960). In the thicker zones, the within-zone variation proved to be larger than the between-zone variation, and chromite compositions in rocks containing both settled olivine and chromite were found to be systematically different from those in rocks containing only settled chromite. As a result, samples collected for determining the lateral and between-zone variation were chosen from a consistent stratigraphic position in the zones (the footwall), and samples containing settled olivine were avoided wherever possible.

The lateral and between-zone changes in composition of chromite was examined simultaneously by plotting values of the major cations at the appropriate places on columnar correlation diagrams, and contouring the values. To simplify interpretation, the columnar diagrams were drawn so that the correlated zones formed horizontal lines. Each of the five major cations of the analyzed chromite from the footwalls of the correlated chromitite zones was plotted individually, and isochemical lines were drawn. None of these major cations, considered individually, shows a simple lateral or between-zone variation. However, two sets of cation values do vary regularly on the diagrams: total iron ($Fe^{2+}+Fe^{3+}$) and oxidation ratio ($Fe^{3+}/Fe^{2+}+Fe^{3+}$). Both values express interaction between the R^{2+} and R^{3+} groups of the chromite.

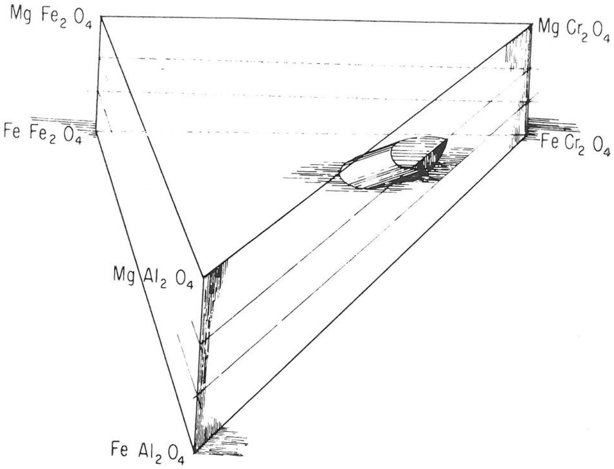

FIG. 4. Compositional range of chromite in the Stillwater Complex.

Between-zone variation The only simple systematic relation between the major cations of the analyzed chromite and the stratigraphic position of the chromitites from which it was separated is shown by total iron. The relation is presented diagrammatically in Fig. 5, where the sum of Fe^{2+} and Fe^{3+} cations/unit cell in the chromite is placed in its appropriate areal and stratigraphic position in the columnar diagrams. In any one stratigraphic section, the values of total iron are greatest in the A zone, decrease regularly to the H zone, and then increase to the K zone. The isochemical lines tend to parallel the layering, emphasizing the strong variation between zones. The A zone values for all sections average 6.1, the B zone values average 5.4, and G zone 4.9, the H zone 4.5 and the K zone 5.5. These averages for cations per unit cell would correspond to total iron expressed as weight per cent FeO of about 27.6, 25.0, 23.2, 21.7 and 25.3 per cent respectively.

Oxidation ratio in the same chromite samples are plotted in Fig. 6. In any one stratigraphic section, the oxidation ratio tends to remain nearly constant, but this relation is not everywhere consistent because the isochemical lines trend at a steep angle across the planes of the zones.

Lateral variation The lateral variation of total iron of the chromite is small, but the systematic depression of isochemical lines in the lower central part of Fig. 5 shows that absolute values are smaller in that area. Considering any given chromitite zone, the values of total iron are about 0.5 cation per unit cell lower in the West Fork section than in sections on either side.

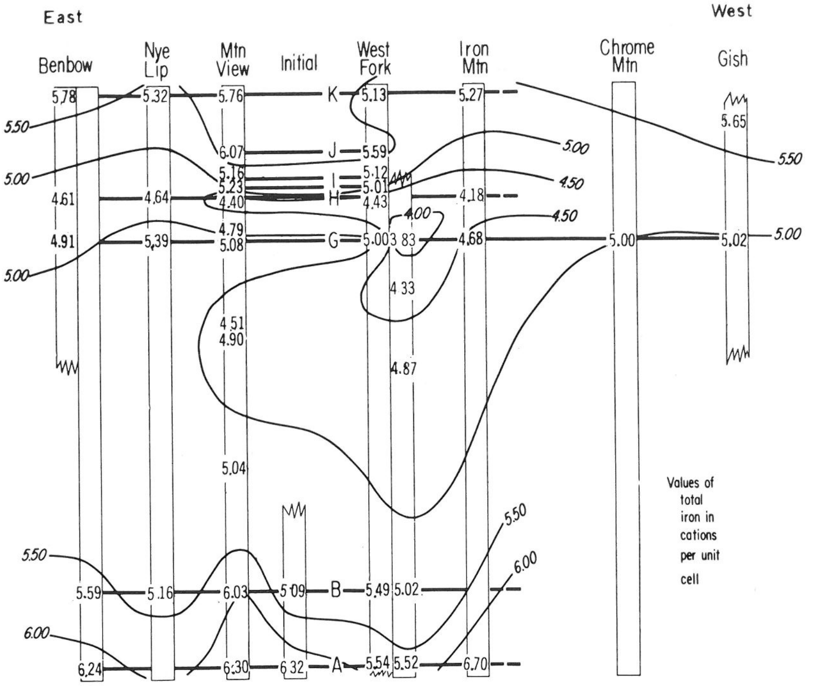

FIG. 5. Columnar diagram of variation of total iron in chromite of the Stillwater Complex.

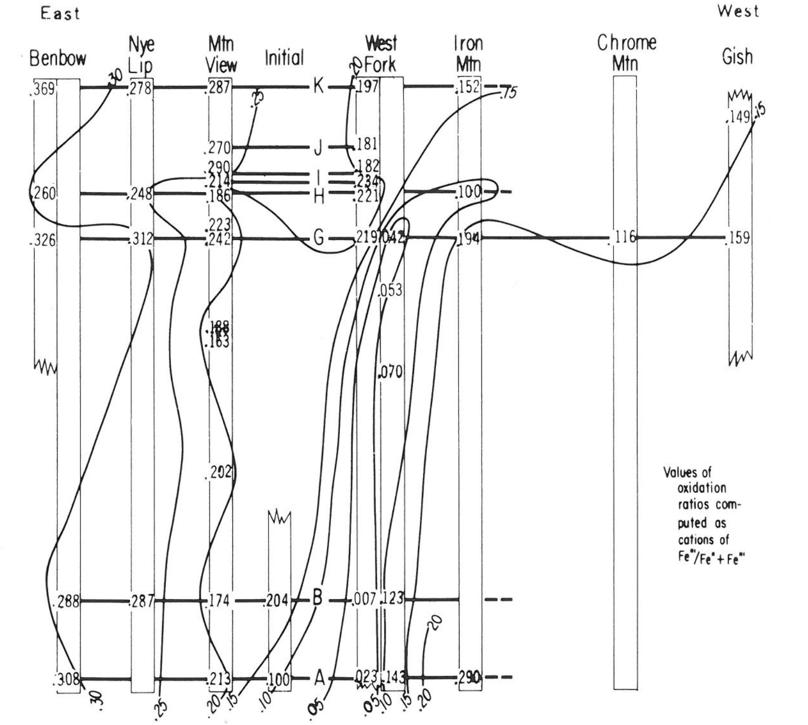

FIG. 6. Columnar diagram of variation of oxidation ratio in chromite of the Stillwater Complex.

A most striking and systematic lateral change in oxidation ratio of the chromite emerges when plotted on the columnar diagrams (Fig. 6). Considering any given chromitite zone, the values of $Fe^{3+}/Fe^{2+}+Fe^{3+}$ are greatest in the chromites at the eastern extremity of the complex, decrease toward the central part, and finally increase again toward the western limit of exposure. The nearly vertical isochemical lines emphasize the lateral nature of the change. The magnitude of this lateral change in oxidation ratio is greater in the lower part of the section than in the upper. In the upper part of the section the average oxidation ratio decreases progressively westward from more than .30 in the Benbow area to less than .10 in the western part of the West Fork section, then increases to more than .15 in the Iron Mountain and Gish areas; in the lower part of the section the oxidation ratio decreases from .30 at both eastern and western ends to less than .05 in the central part of the complex.

Inasmuch as the total numbers of both bivalent and trivalent cations per unit formula of balanced chromite are fixed, this combination of nearly constant total iron and changing oxidation ratio of chromite along any given chromitite zone accounts for the complex variations of the individual cations. Fe^{2+}, for example, is low along the H zone chromitite where total iron is low, but is also low in the upper and lower zone chromitites of the Benbow area where, although total iron is high, much of it occurs in the trivalent form. The highest values of Fe^{2+} occur in the lower part of the Initial and West Fork areas, where total iron is high and oxidation ratio low. Mg^{2+} shows a reciprocal pattern. Of the trivalent cations in the chromites, Fe^{3+} is low in the central part of the complex where oxidation ratio is low, but it is also low along the G and H zone chromitite where total iron is low, even in areas where the oxidation ratio is high. The amount of Cr^{3+} in the chromite shows a nearly reciprocal relation with Fe^{3+}, so that high Cr^{3+} chromite occurs in the central part of the complex and also laterally along the G and H zone chromitites. As a consequence of these changes, the $Cr^{3+}/Fe^{2+}+Fe^{3+}$ ratio of the chromite has a pattern almost reciprocal to that of $Fe^{2+}+Fe^{3+}$.

The existence of this regular pattern should permit the prediction of the composition of chromite anywhere in the complex. As a check on the extrapolation of isochemical lines between sampled sections, several previously published analyses of Stillwater chromites from various localities in the Peridotite member (Stevens, 1944, p. 12–13 and 22; Howland *et al.*, 1949, p. 76; Howland, 1955, p. 109) were entered on the diagrams. About 20 of these older analyses were of samples collected near the footwalls of massive chromitite layers. Values of total iron and, where determined, oxidation ratio of these samples fell within the established pattern.

The significance of the lateral and between-zone variations in iron content and oxidation state in the chromite, can, at this time, be outlined only in a very general way. It should be noted at the outset, however, that it is very unlikely that the observed differences in composition could have originated after the crystallization of the mineral. Oxidation or reduction of iron in chromite destroys the charge balance of the mineral and cannot be accomplished without large gains or losses in other major cations by diffusion or without the generation of another oxide phase. Alteration of chromite has been described by a number of authors and is evidenced in three ways:

(1) exsolution of an α (R_2O_3) phase (de Wijkerslooth, 1942, p. 267–289); (2) presence of a secondary spinel phase (Miller, 1953, p. 1134–1147); or (3) development of γ (defect) structures in the chromite (Rait, 1946, p. 191–192).

The analyzed Stillwater chromites are internally homogeneous, closely packed, for the most part euhedral, and are the only oxide phase present in the rocks. Careful inspection in reflected light has shown that no significant amounts of R_2O_3 or other spinel phases are present, and the cation abundances calculated from the analyses show that vacant lattice positions required by γ structures do not exist. Furthermore, it is unlikely that any post-magmatic alteration could have occurred without affecting the fresh olivine with which the chromite is associated. The observed variations in chromite compositions are therefore believed to represent differences in magmatic conditions during the crystallization of the chromite.

It has been proposed elsewhere (Jackson, 1961, p. 81–99) that the massive chromitites of the Stillwater Complex are accumulations of chromite crystals that grew near the floor of the magma chamber and settled in relatively stagnant magma without a great deal of lateral transport. Chromites at the bases of individual chromitite zones, therefore, are believed to have crystallized simultaneously over a wide area of the floor of the complex. Presumably lateral changes in the composition of chromite represent differences in temperature, pressure, or composition of the magma above a particular part of the floor at a given instant of time. The principal lateral changes in chromite composition are a large decrease in the proportion of ferric iron and a small decrease in total iron toward the central part of the complex. If the oxidation ratio of the chromite reflects that of the magma from which it crystallized, then it is likely that a lateral oxygen partial pressure gradient (or more properly an oxygen fugacity gradient) existed during most of the crystallization and accumulation of the Peridotite member. Such a gradient might be related to the convection cell pattern, to the position of feeders, or to extraction of water from intruded sediments at the margins of the intrusion. The presence of small amounts (0–1.6 volume per cent) of interprecipitate phlogopitic mica in the chromitites suggests that water was present in the magma during crystallization and accumulation of the chromite. Attempts to correlate amounts of mica in chromitites with oxidation ratio of chromite from them are complicated by the variable amounts of interprecipitate material in the rocks and by the difficulty of accurately measuring small amounts of the poikilitic interprecipitate minerals. Work on these measurements is continuing.

It is apparent from the law of superposition that the upper chromitite zones were successively crystallized and deposited on the lower. Thus variations in composition of chromite between zones should reflect changes in magma temperature, pressure, or composition with time. The principal between-zone changes in chromite composition is a steady decrease in total iron to the H zone chromitite followed by a steady increase in iron to the K. Normally, one would expect that total iron would be lowest in the A zone chromite and that it would increase upward for two reasons:

1. The iron end members of the spinel solid solution series have the lowest melting points and should increase in the chromite with time as heat is extracted from the crystallizing system; and
2. The magma should become residually enriched in iron with time due to fractionation of large quantities of magnesium-rich olivine and orthopyroxene.

The opposite of this expected relation in the lower zones may be due to unknown complications of fractionation paths in the chromite fields of the chemical systems involved, or to failure of the magma to reach complete thermal equilibrium during crystallization of the lower part of the Peridotite member, or to successive injection of increasingly iron-poor magmas. Studies of the vertical variation in iron content of olivine and bronzite may allow a choice between these hypotheses, and work on these minerals is in progress.

OTHER STRATIFORM-TYPE CHROMITES

Comparison of variations in chromite composition in the Stillwater Complex with those of chromite from other stratiform intrusions is hampered by incomplete stratigraphic control and by a general scarcity of trustworthy chemical analyses. Enough information is available, however, to suggest that the Stillwater variation may not be unique.

Worst (1958) reports 11 complete analyses of clean chromite from 9 chromitite layers of known stratigraphic position in the Hartley area of the Great Dyke of Southern Rhodesia. The samples were collected from both margins of the Dyke along a strike length of more than 50 miles, and were obtained by channelling the full width of the layers. Worst's seam numbers, arranged in their proper sequence, his stratigraphic intervals between the seams, and his reported FeO and Fe_2O_3 contents of the chromite from the seams are listed in Table 1. I have recalculated the FeO and Fe_2O_3 values of the analyses to both total iron as FeO and to $Fe^{2+}+Fe^{3+}$ per unit cell. Inspection of the table shows that although FeO and Fe_2O_3, considered individually, show no particular vertical order, the values of total iron have a variation most similar to that in the Stillwater Complex. With the single exception of analysis 10, total iron decreases regularly from seam 9 to seam 7, then increases regularly to seam 1. The oxidation ratio of the chromite, also calculated in Table 1, shows only that a considerable range of values exists, as would the Stillwater samples if collected at irregular intervals along the strike of the intrusion.

TABLE 1. STRATIGRAPHIC VARIATION OF IRON CONTENT OF CHROMITE FROM THE HARTLEY AREA, GREAT DYKE OF SOUTHERN RHODESIA

Seam No.[1]	Stratigraphic interval[1] (feet)	FeO in clean chromite[1] (Wt. per cent)	Fe_2O_3 in clean chromite[1] (Wt. per cent)	Total iron as FeO[2] (Wt. per cent)	$Fe^{2+}+Fe^{3+}$[2] (cations per unit cell)	Oxidation ratio[2]
1[3]	0	13.33	9.90	22.24	4.81	.401
2[3]	100	12.06	8.74	19.92	4.18	.395
3	460	Not analyzed		—	—	—
4	1,730	9.98	9.19	18.25	3.91	.453
5	660	11.82	4.56	15.92	3.37	.258
6	390	8.88	6.76	14.96	3.17	.407
7	310	9.78	5.12	14.39	3.07	.319
8	440	11.29	4.71	15.53	3.33	.273
9	380	8.80	8.15	16.13	3.44	.453
10	420	10.86	4.99	15.35	3.30	.291

[1] From Worst (1958, Tables 13 and 15).

[2] Calculated from Worst's data.

[3] Chromite analyses 1 HW and 2F are apparently from olivine chromites, and are omitted from this tabulation.

In the Bushveld Complex, about 60 analyses of clean chromite from systematically collected chromitites have been published by deWet (1952a, 1952b, 1956). As the analyses are not complete, balance cannot be checked, but both FeO and Fe_2O_3 were determined.

In the western belt of the Bushveld Complex, chromite from a sequence of chromitites in the Swartkop area was analyzed with the results shown in Table 2. Inspection of the table shows that both FeO and Fe_2O_3 decrease, then increase stratigraphically upward, but the minimums are reached in different seams. Total iron as FeO decreases from about 25 per cent in the Compound seam to about 20 per cent in the Intermediate seam, stays about the same in the New seam, then increases to about 26 per cent in the Magazine seam. Although the Upper group and Merensky Reef seams were not sampled in this area, it is probable from analyses in other areas that they are richer in total iron than the Magazine. Thus it is not improbable that the chromite compositional variation in this area is similar to that observed in the Stillwater Complex.

In the eastern chromite belt of the Bushveld Complex, deWet made no systematic collections through Lower and Middle group seams at any one locality. Averages of total iron in clean chromite from various localities of the eastern belt show that the Lower group chromite averages about 26 per cent total iron as FeO, Middle group chromite averages about 30 per cent, and one sample from the Merensky Reef gave 36 per cent (deWet, 1952a, Tables I, II, III and 1956, Table IV). There is no suggestion of a reversal.

TABLE 2. STRATIGRAPHIC VARIATION OF IRON CONTENT OF CHROMITE FROM THE SWARTKOP-SCHILPADNEST AREA, WESTERN BUSHVELD COMPLEX

Seam[1]	Group[1]	No. of analyses[1]	FeO in clean chromite[2] (wt. per cent)	Fe_2O_3 in clean chromite[2] (wt. per cent)	Total iron as FeO[2] (wt. per cent)	Oxidation ratio[3]
Magazine	Lower or Middle	Average of 9	17.0	9.6	25.7	.337
New	Lower	Average of 4	14.0	6.9	20.1	.307
Intermediate	Lower	Average of 4	11.9	9.3	20.3	.413
Compound	Lower	Average of 3	15.3	10.5	24.8	.382

[1] From deWet (1952b, p. 1–10).
[2] Average from deWet (1952b, Tables V, VI, VII, VIII).
[3] Calculated from deWet's data.

TABLE 3. LATERAL VARIATION OF IRON CONTENT OF CHROMITE FROM THE LOWER GROUP, EASTERN BUSHVELD COMPLEX

	Location		Steelpoort seam and probable equivalents		Steelpoort leader seam	
			Total iron as FeO in clean chromite[1] (wt. per cent)	Oxidation ratio[2]	Total iron as FeO in clean chromite[1] (wt. per cent)	Oxidation ratio[2]
North	Jadglust	333	25.4	.362	25.9	.383
↑	Winterveld	343	24.8	.387	24.9	.380
	Mecklenburg	371	26.4 (avg. of 2)	.352	25.9	.322
	Clapham	364	24.1	.319	24.4	.388
	Twyfelaar	172	25.7	.322		
	Mooihoek	147	26.9	.401		
	Hendriks Plaats	357	26.5	.524	26.4	.449
	Winterveld	424	25.4 (avg. of 2)	.388	25.2	.391
	Grootboom	186	26.1	.210	26.9	.218
	Annex Grootboom	473	25.9	.243		
↓	Tweefontein	35	25.0	.244		
South	Thorncliffe	217	25.1	.258		

[1] From deWet (1952a, Tables I and II).
[2] Calculated from deWet's data.

DeWet also investigated the lateral compositional variation of chromites from the Steelpoort seam and its probable Lower group equivalents along a strike distance of nearly 60 miles in the eastern belt. His results are summarized in Table 3. The chromites of the Steelpoort seam and of the Steelpoort leader seam just above it have nearly identical total iron contents along the strike; deviation from the average in each is only ±5 per cent. The oxidation ratios of both seams, on the other hand, appear to decrease regularly from the northernmost exposure to a place near Clapham 364, then to increase strongly to a place near Hendriks Plaats 357, and finally to decrease toward the south; deviation from the mean is almost 40 per cent. This situation would seem to be similar to that found in the Stillwater Complex, where total iron in chromite is nearly constant along the zones, but where oxidation ratio shows wide systemic variations.

CONCLUSION

The chromitite zones of the Stillwater Complex have been correlated by field mapping. Chemical analyses of chromite from these zones show that the variation in composition within single zones is large; therefore samples from consistent stratigraphic positions and of given rock types must be compared to determine lateral and between-zone compositional variations. When such samples are compared, a simple variation in total iron between zones is observed, such that $Fe^{2+}+Fe^{3+}$ in the chromite decreases from the lower zones toward the middle zones, then changes, and increases regularly through the upper zones. At the same time, a lateral variation in oxidation ratio of the iron in the chromite is observed such that the ratio of Fe^{3+} to total iron decreases from the eastern and western margins of the complex toward the central part, particularly in the lower chromitite zones. The nature and regularity of the changes permit the prediction of chromite composition within and between the sampled sections.

These systematic changes in composition are believed to reflect differences in magmatic conditions during the crystallization of the chromite. The lateral variation probably represents a persistent oxygen partial pressure gradient across the intrusion. The variation between zones probably represents an early reversal in the trend of magma composition or in crystallization temperature of the chromite during differentiation of the intrusion.

Examination of published chromite analyses from the Bushveld Complex and from the Great Dyke suggests that similar variations in chromite composition may be present in these intrusions.

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