

COMPOSITION OF LIQUIDUS CHROMITE FROM THE 1959
(KILAUEA IKI) AND 1965 (MAKAOPUHI) ERUPTIONS
OF KILAUEA VOLCANO, HAWAII¹

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ABSTRACT

The compositional variation of liquidus chromite is defined for the tholeiitic lavas of the 1959 Kilauea Iki and 1965 Makaopuhi eruptions. A trend of decreasing Cr_2O_3 is accompanied by decreasing MgO and Al_2O_3 , and by increasing FeO, Fe_2O_3 , TiO_2 , and V_2O_5 . These trends are observable in rapidly quenched basaltic pumices, but are significantly modified during subsequent crystallization of the host basalt. Correlation between the composition of chromite and olivine suggests that mutual equilibrium was attained and that compositions were preserved by rapid quenching. Temperatures of apparent equilibrium for chromite-olivine pairs were determined as 2000–2300°C using an expression derived by Jackson (1969) and are far in excess of the eruption temperatures (1200° to 1225°C). The composition of the olivine controlling the composition of the 1959 and 1965 eruptions are, respectively, $\text{Fa}_{23.7}$ and $\text{Fa}_{25.4}$. The weight percent of chromite in olivine is 1.2 in the 1959 lavas and 1.5 in the 1965 lavas.

INTRODUCTION

Tiny idiomorphic crystals of chromite are of ubiquitous occurrence in Kilauean lavas. They are commonly included in or attached to phenocrysts of magnesian olivine and, in rapidly cooled lavas, they are present in addition as isolated grains in basaltic glass. The chromite and associated olivine are the earliest phases to crystallize from Kilauean tholeiitic magma. With the exception of data in Evans and Moore (1968), the composition of chromite from Kilauea has not previously been determined. In order to study the changes in composition of chromite that occur at temperatures below those corresponding to its primary field of stability, it is necessary to establish the primary compositional trends. Such trends can reasonably be expected to be qualitatively true for liquidus chromite in other basaltic magmas of comparable composition and state of oxidation.

As first illustrated by Powers (1955) and subsequently by Murata

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and Richter (1966a, b) and Wright (1971), the variation in bulk chemistry of Kilauean lavas from many eruptions is controlled by the addition or removal of olivine. When plotted on MgO-variation diagrams, the chemical data define olivine control lines which extrapolate to the composition of the olivine (plus attached or included chromite). Knowledge of the chromite chemistry is necessary to fix the average amount of chromite in the olivine and also to determine correctly the composition of olivine (without chromite) that controls the chemistry.

The chemical composition of liquidus chromite is a significant factor in an understanding of the conditions governing the early stages in the crystallization of basaltic magmas under low total pressure. The concentrations of some constituents of liquidus spinel are very sensitive to the fugacity of oxygen, for example (Hill and Roedder, 1967; Ulmer, 1969). Further, there seems to be some suspicion as to the influence of post-cumulus events on the composition of primary precipitate chromite and coexisting silicate in stratiform ultramafic intrusives (Irvine, 1967; Jackson, 1969; Cameron, 1969, 1970; Cameron and Desborough, 1969). Therefore, it is a logical step to establish the composition of volcanic chromite precipitated from comparable mafic magma, particularly that of Kilauea, whose properties are already rather well known.

Electron microprobe analyses have been conducted on chromites and associated olivine phenocrysts from the 1959 eruption in Kilauea Iki pit crater and two from the 1965 eruption in Makaopuhi pit crater (Table 1). One sample from each eruption was collected from the surface of a rising lava lake. The rest are pumice.

COMPOSITION OF CHROMITE

The major element contents of rocks whose chromites are studied here are given in Murata and Richter (1966a, Table 1), and in Wright *et al.* (1968, Table 6). The Cr_2O_3 -contents of the rocks are given in Table 2. Average compositions of chromite from Kilauea Iki and Makaopuhi are also given in Table 2 and individual grain analyses are recorded on Figure 1.

The grain-sizes of the chromites are typically 20 to 40 μm in diameter; chromites exceeding 100 μm in section are unusual. None of those analyzed was less than 12 μm in cross section. The low analysis totals in Table 2 are undoubtedly due to the inclusion of grains less than about 25 μm in mean diameter. A plot of analysis total versus grain-size clearly showed a loss in X-ray intensity in small grains relative to large grains (and, of course, relative to the large grains of analyzed chromites used for standardization). The loss is estimated to be

about 1 percent relative to the amount present for 20 μm diameter grains, and $1\frac{1}{2}$ percent for 10 μm grains. It is principally in Cr and Fe, and the loss arises from the absence in small grains of fluorescence-excited $\text{CrK}\alpha$ and $\text{FeK}\alpha$ from areas distant from the point of electron impact.

Many of the analyzed chromites were totally enclosed in olivine, many were partially in olivine and partially in glass, and some were entirely enclosed in glass. Microprobe analyses failed to show any systematic differences between chromite in olivine and chromite in glass, save possibly for the greater grain-to-grain uniformity in the latter case.

Chromites from Kilauea Iki are high in Cr_2O_3 (40 to 46 percent), whereas those from Makaopuhi are slightly lower (35 to 43 percent Cr_2O_3). The variation in chrome content is accompanied by systematic variation in the other constituents; decrease in Cr_2O_3 is correlated with decrease in MgO and Al_2O_3 and increase in FeO, Fe_2O_3 , TiO_2 , and V_2O_5 (Fig. 1). These variations are between grains rather than within

TABLE 1. DETAILS OF ANALYZED SAMPLES.

Analyzed Samples			
Eruption	Sample No.	Quenching Temperature °C ¹	Type of Material
Kilauea Iki (1959)	S-5	1225°	Ejected pumice
		MgO (glass) = 10.2%	
	S-15	n.d.	Ejected pumice
	S-21	n.d.	Ejected pumice
	S-20	n.d.	Sample collected from lake, quenched in air.
Makaopuhi (1965)	M-1	1200°	Ejected pumice
		MgO (bulk) = 8.3%	
	M-18	1170°	Sample collected from lake in ceramic, quenched in water

¹ Eruption temperature of S-5 inferred from the iron/magnesium ratio in separated glass (Murata and Richter, 1966a, Table 4, and Tilley, Yoder and Schairer, 1966). Temperature of M-18 is estimated from thermocouple measurement at the time of collection. Eruption temperature of M-1 is estimated to be 5° cooler than the liquidus temperature estimated from the iron/magnesium ratio of the bulk rock and the calibration of Tilley, Yoder and Schairer (1966). See Wright and others (1968, Table 5) for additional data on temperatures of Kilauea eruptions.

n.d. not determined.

Table 2.--Average Composition of Chromites and Cr_2O_3
content of the chromite-bearing lavas

	Kilauea Iki				Makaopuhi	
	S-5	S-15	S-21	S-20	M-1	M-18
$\frac{1/}{\text{SiO}_2}$.09	.09	.10	.08		
TiO_2	2.30	2.28	2.47	2.32	3.15	3.20
Al_2O_3	13.5	13.3	13.1	13.7	12.85	13.1
Cr_2O_3	43.3	43.8	42.4	43.0	38.4	38.5
Fe_2O_3 ^{2/}	10.81	10.70	11.17	10.35		
FeO ^{2/}	15.27	15.50	17.04	17.46	25.1 Fe	25.6 Fe
MnO	.17	.17	.19	.19		
MgO	13.2	13.1	12.0	12.0	10.7	10.2
CaO	.01	.05	.13	.02		
ZnO	.10	.11	.12	.12		
NiO	.16	.16	.15	.15		
V_2O_3	.19	.20	.20	.22		
	99.10	99.46	99.07	99.61		
No. of grains	7	9	5	20	14	15
Av. grain cross-section (μm)	29	30	21	31	not recorded	not recorded
$\frac{3/}{\text{Cr}_2\text{O}_3}$ (rock)	.20	.16	.17	.14	.056	.059

^{1/}Tests involving chromites 150 μm across showed that boundary fluorescence contributes no more than .01% SiO_2 further than 4 μm from the grain boundary.

^{2/} FeO and Fe_2O_3 computed from total iron assuming perfect spinel stoichiometry.

^{3/} Cr_2O_3 analyses are on splits of the same powder on which the major oxide analysis was made (Murata and Richter, 1966a, Table 1, Wright, et al., 1968, Table 6 and unpublished data). Method: average of colorimetric and atomic absorption. Analyst: V.E. Shaw, U.S. Geological Survey.

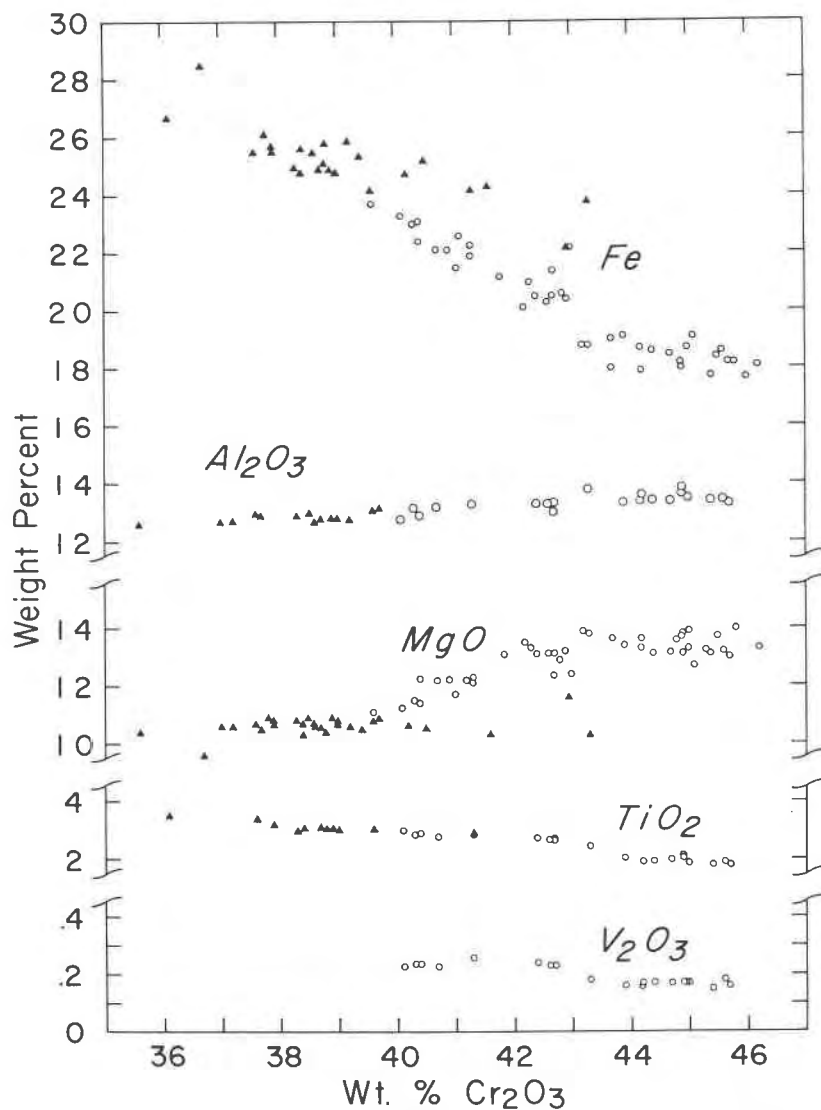


FIG. 1. Compositional trends in individual chromites from Kilauea Iki (circles) and Makaopuhi (triangles). Samples K.I. S-20 and M-18 omitted (see Figure 4). Note that Al, Ti, and V were determined during a second microprobe run after Mg and Fe.

grains of chromite; zoning of chromite is only detectable in the larger grains and even in these it is often trivial in extent. Thus, the profile in Figure 2 is atypical, whereas that in Figure 3 is typical.

The compositional trends within the Kilauea Iki and Makaopuhi suites are similar and more-or-less continuous with one another, although the Makaopuhi chromites with higher Cr_2O_3 possess lower MgO than the Kilauea Iki chromites of comparable Cr_2O_3 content. The increase

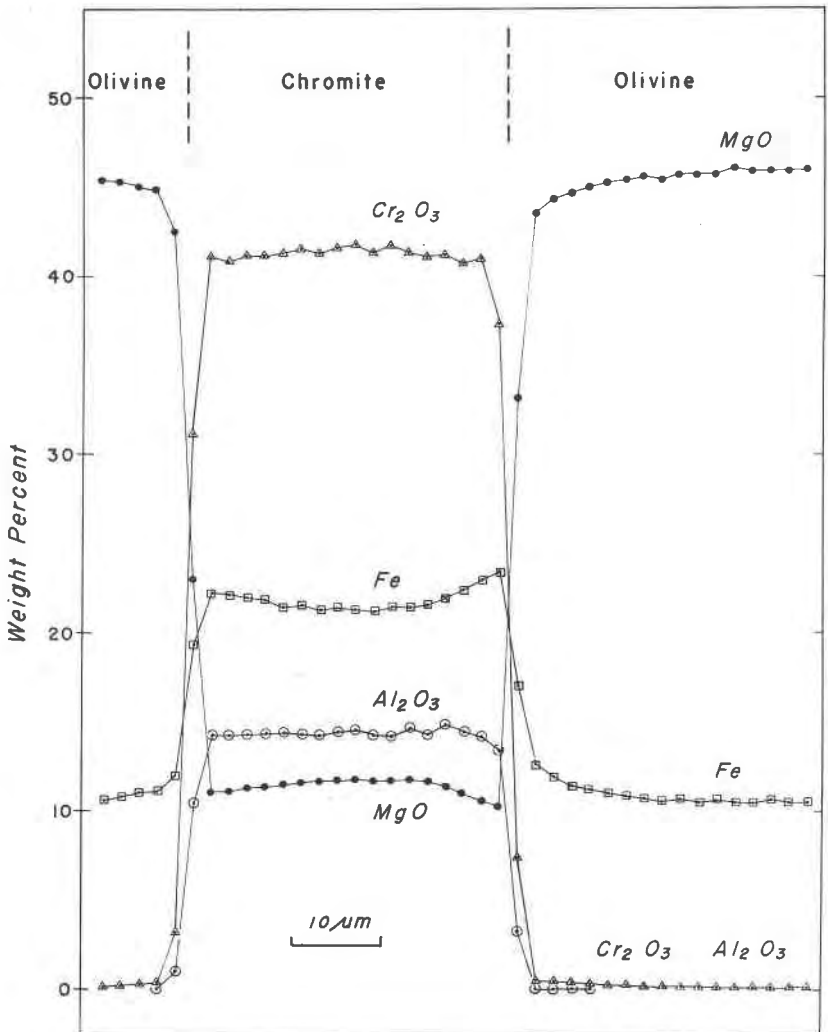


FIG. 2. Profile across a slightly zoned chromite inclusion (S-20, grain 4).

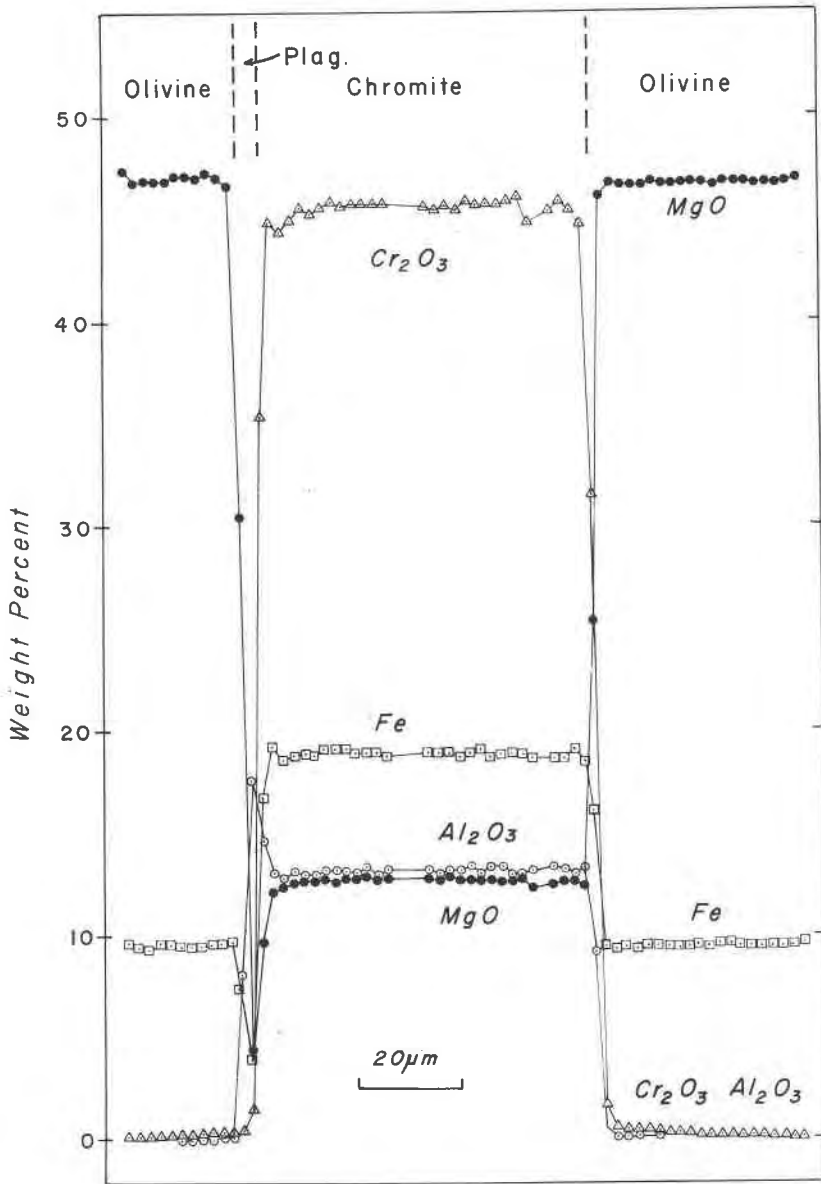


FIG. 3. Profile across a typical homogeneous chromite grain (S-20, grain 7).

in TiO_2 with decrease in Cr_2O_3 is comparable to the inferred pre-eruption trend of chromite in the prehistoric Makaopuhi lava lake (Evans and Moore, 1968). The trend of increasing Al_2O_3 with decreasing Cr_2O_3 for the prehistoric Makaopuhi chromites, however, is not confirmed by the present study (Fig. 1). Expressed in molecular terms, roughly equal amounts of magnetite and ulvöspinel are gained at the expense of chrome-spinel and, to a lesser degree, aluminous spinel. These changes correspond to falling temperatures and increased crystallization of chromite and olivine, accompanied, presumably, by decreasing f_{O_2} .

Two samples (Kilauea Iki S-20, and Makaopuhi M-18) do not show the excellent inter-element trends exhibited by the other chromites (Fig. 4). Both samples were collected on ceramic combustion tubes from a rising lava lake and were quickly quenched either in air or water. The temperature of collection was several tens of degrees below the eruption temperature and in both samples a poorly understood process of alteration of the chromite appears to have been initiated, analogous to that observed in the prehistoric Makaopuhi lava lake (Evans and Moore, 1968, pp. 103-106). Even though many are enclosed by olivine phenocrysts, the early liquidus chromites are clearly highly sensitive to subsequent events, and it seems that their unaltered preservation can only be assured in basaltic pumice quenched by ejection into the air.

One chemical property that seems to distinguish Kilauean chromite from that of stratiform ultramafic bodies (Stevens, 1944; Howland, 1955; Cameron and Emerson, 1959; Worst, 1960; Thayer, 1964, 1969; Cotterill, 1969; Bichan, 1969; Jackson, unpublished analyses of Stillwater chromite) is the relatively high TiO_2 of the former. Gunn, et al (1970) report a chromite with only 1.2 percent TiO_2 from an Indian Ocean oceanite, which suggests that Kilauean chromite may owe its richness in TiO_2 to the magma's relatively high TiO_2 content (Murata and Richter, 1966a). In addition, chromite precipitating at a slightly later stage from a basaltic magma is likely to contain higher concentrations of the ulvöspinel molecule.

The relatively high ratio of $\text{Fe}_2\text{O}_3/\text{FeO}$ for Kilauean chromite (Table 2) may be more apparent than real, since it was determined indirectly, by calculation. Analyses of most, but not all, ultramafic stratiform chromites show lower proportions of ferric iron.

CHROMITE-OLIVINE RELATIONSHIPS

Olivine in the Kilauea Iki samples studied varies in $\text{Fe}/(\text{Fe} + \text{Mg})$ ratio from .12 to .16; that from Makaopuhi ranges from .154 to .184 (Table 3). A close correlation was found to exist between the composi-

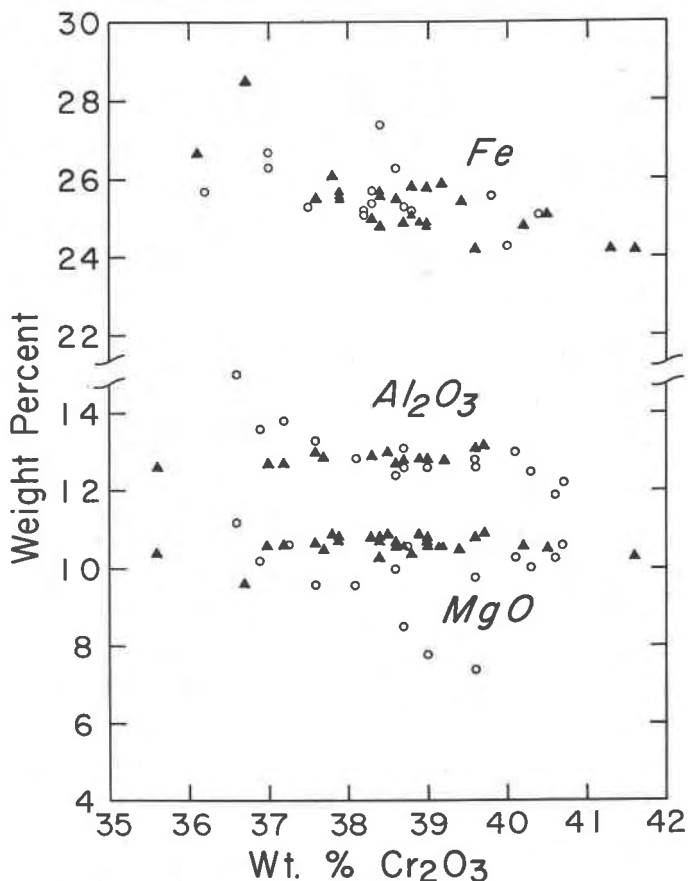


FIG. 4. Compositional trends for Makaopuhi chromite M-1 (triangles), compared with poor trend for M-18 (circles).

tion of chromite and that of the host olivine phenocryst or olivine phenocryst to which the chromite was attached; the more Mg-rich olivines are associated with the more Mg- and Cr-rich chromites. The relationship between $Fe/(Fe + Mg)$ in olivine and $Fe^{2+}/(Mg + Fe^{2+})$ ¹ in chromite from Kilauea (Fig. 5) almost certainly extends to the more iron-rich Makaopuhi olivines, although Fe^{2+} was not separately evaluated for the Makaopuhi chromites because full analyses were not

¹ The determination of $Fe^{2+}/(Mg+Fe^{2+})$ and Fe^{3+} in chromite is subject to the cumulative errors in the determination of all the other constituents, and to the assumption of exact spinel stoichiometry. Variation in the relative peak heights of $FeL\alpha$ and $FeL\beta$ in chromite was found to be insensitive to the ratio Fe^{3+}/Fe^{2+} .

done. Analogous relationships have been demonstrated in ultramafic intrusives (Jackson, 1969; Cameron, 1970). However, the conditions governing the evident equilibrium partitioning of elements between olivine and chromite in Kilauean basaltic pumice are much less ambiguous than in the plutonic rocks. During intratelluric crystallization over a narrow range of falling temperature, olivine and chromite underwent gradual changes in chemistry in response to the changing liquid composition. Successive crops of crystals appear to have been in good mutual equilibrium. Subsolidus reequilibration between olivine and chromite is ruled out by the quench origin of the pumice. Progress towards subliquidus reequilibration between the two solids, isolated from liquid, during the minor temperature decline before quenching, may have taken place but there is no evidence for it.

Table 3. Derivation of olivine - chromite apparent equilibration temperatures, using the equation of Jackson (1969)

		Olivine Fe/(Mg+Fe)	Fe ²⁺ /(Mg+Fe ²⁺)	Chromite			K _D Mg-Fe ²⁺	T ^o C
				α	β	σ		
S-21	1	.123	.394	.615	.267	.119	4.64	2134
	2	.123	.387	.615	.270	.115	4.50	2177
	3	.154	.471	.560	.273	.166	4.90	2045
	4	.154	.482	.564	.268	.168	5.12	1982
	5	.153	.475	.564	.269	.167	5.01	2019
S-20	1 Center	.131	.377	.580	.271	.150	4.01	2490
	1 Edge	.131	.426	.591	.272	.138	4.92	2024
	4	.148	.482	.568	.296	.136	5.37	1799
	5	.148	.480	.567	.294	.139	5.32	1821
	6	.148	.469	.570	.294	.137	5.09	1891
	6A	.144	.468	.564	.298	.138	5.22	1836
	7 Center	.127	.412	.615	.265	.120	4.82	2069
	7 Edge	.127	.414	.616	.266	.118	4.86	2050
	8	.128	.411	.615	.265	.120	4.76	2091
	9	.128	.440	.619	.259	.123	5.35	1910
	11	.140	.446	.588	.277	.135	4.94	1995
	12	.140	.440	.589	.275	.136	4.82	2045
	13	.126	.421	.575	.289	.136	5.05	1920
	14	.126	.429	.576	.291	.134	5.20	1863
15	.146	.464	.595	.268	.137	5.05	1987	
16	.146	.468	.582	.269	.148	5.14	1961	
17	.146	.456	.588	.266	.146	4.90	2056	
18	.141	.474	.562	.298	.140	5.49	1762	
19	.135	.428	.570	.284	.146	4.80	2032	
S-5	1	.146	.450	.571	.274	.154	4.78	2080
	2	.146	.443	.564	.273	.164	4.64	2155
	3	.120	.371	.602	.274	.124	4.33	2256
	4	.120	.365	.597	.274	.129	4.21	2329
	5	.120	.373	.585	.278	.137	4.36	2242
	6	.126	.368	.600	.271	.129	4.03	2442
	7	.126	.381	.592	.267	.141	4.27	2332
S-15	1	.132	.407	.581	.270	.149	4.52	2206
	2	.132	.404	.581	.271	.148	4.46	2231
	3	.126	.390	.604	.270	.126	4.44	2221
	4	.126	.391	.602	.271	.128	4.46	2207
	5	.126	.382	.596	.270	.134	4.29	2305
	5A Center	.127	.391	.609	.267	.124	4.42	2240
	5A Edge	.127	.385	.601	.269	.130	4.30	2299
6	.144	.438	.584	.265	.151	4.63	2176	
7	.134	.404	.578	.270	.152	4.39	2273	

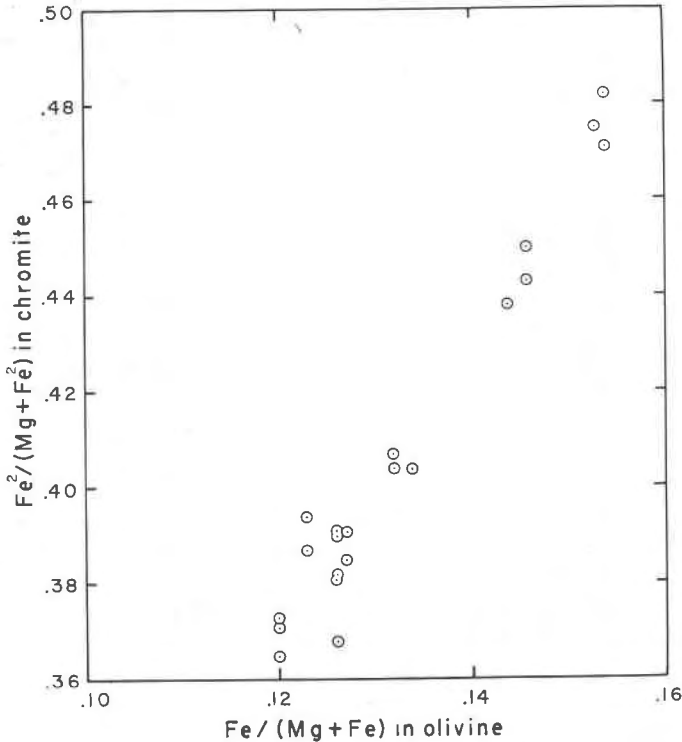


FIG. 5. Correlation between $\text{Fe}^{2+}/(\text{Fe}^{2+} + \text{Mg})$ in chromite and associated olivine from Kilauea Iki pumices. Sample S-20 omitted (p. 224).

In any event, it is instructive to apply Jackson's expression (1969, p. 63, equation 9) for the temperature of equilibrated olivine and chromite (Table 3). Apparent temperatures range from over 2300°C for high Cr, high Mg pairs to less than 2000°C for low Cr, low Mg pairs. Whereas the direction of falling temperature is consistent with the temperature change that would be inferred from the $\text{Fe}/(\text{Fe} + \text{Mg})$ ratio of the olivine, the values are obviously too high by about 1000°C ¹. Liquidus temperatures for samples of the 1959 Kilauea Iki eruption of up to 1225°C can be inferred from a combination of the experimental data of Tilley *et al.* (1966) and the observed iron/magnesium ratios of separated Kilauea Iki glasses (Murata and

¹Temperatures derived in Table 3 are clearly sensitive to errors in Fe^{2+} and Fe^{3+} ; they would only be 700°C lower if *all* the iron were in the ferrous state, and roughly 400°C lower if the ratio $\text{Fe}^{2+}/\text{Fe}^{3+}$ were assumed to be comparable to Stillwater chromite.

Richter, 1966a)—the temperature derivation is given by Wright *et al.*, 1968, Table 5. S-20 is the only Kilauea Iki sample obtained from the lava lake; this extended the cooling history of the sample sufficiently to give generally lower and slightly less consistent "temperatures" than the other Kilauea Iki samples.

Using the same geothermometer, Jackson (1969) obtained temperatures of 900°C to 1400°C (mostly 1100°C to 1300°C) for the chromitite zones of the Stillwater Complex. Similar values (1098° to 1335°C) were found by Loney, *et al.* (1971) for the alpine-type peridotite at Burro Mountain, California, and slightly higher values (1200°C to 1410°C) for high-pressure peridotites in SW Oregon were determined by Medaris (Ms).

Irvine (1967, p. 85) has argued that, to obtain the best estimate of the initial temperature of crystallization of assemblages of chromian spinel and orthopyroxene or olivine in layered intrusions, one should use partition coefficients that compare the compositions of the minerals as they occur at maximum concentration in adjacent layers, rather than as they occur in adjacent grains in a sample of one rock type. If this procedure is followed for the Stillwater chromitite layers, using data from Jackson (1969, Tables 1 and 2), a range of "temperatures" from 1450°C to 2020°C is obtained. The result is that the Stillwater and Kilauea Iki "temperatures" now look more similar, as one might reasonably expect them to do.

It is suggested that the equation of Jackson cannot at present be applied to obtain temperatures of formation for terrestrial volcanic chromites, analyzed by the microprobe. The reasons for the lack of success are probably one or more of the following: 1) the uncertainty in the calculation of ferrous and ferric iron assuming a spinel stoichiometry, 2) the high TiO₂ content of volcanic chromite which makes it imperative that the activity of TiO₂ (or ulvöspinel) be considered in deriving the equations, 3) deficiencies in Jackson's temperature expression (some of the possible shortcomings in the derivation were discussed in his work (Jackson, 1969)).

OLIVINE-CHROMITE CONTROL OF LAVA COMPOSITION

The chemistry of the 1959 eruption has been described by Murata and Richter (1966a). The composition of olivine (+ chromite) has been derived from extrapolation to high-MgO contents of the olivine control lines defined by the composition of the erupted lavas (Murata and Richter, 1966b). Data of the present paper along with new Cr₂O₃ determinations of bulk lava compositions have been used by T. L. Wright and D. B. Jackson (ms. in prep.) to refine Murata and Richter's

olivine composition and to determine the weight percent ratio of chromite to olivine + chromite. The average olivine composition so determined is $Fa_{13.7}$ compared to the previously determined value of $Fa_{13.6}$ (Murata and Richter, 1966b, p. 196). The percentage of chromite varies from 1.0 to 1.4, averaging 1.2.

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