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THE MINERAL CHEMISTRY OF THE INNISFREE METEORITE

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Abstract

The Innisfree meteorite is an LL-type, possibly genomict, brecciated chondrite. Electron-microprobe analyses of plagioclase, orthopyroxene, clinopyroxene, olivine, chromite, ilmenite, merrillite (= whitlockite), apatite, troilite, kamacite, taenite and devitrified chondrule glass are presented. Very wide variations in Ni/Fe and Ni/Co ratios in the metal phases are contrasted with patterns of variation in a number of other chondrites. Together, these variations tend to support an autometamorphic model for the equilibration of chondrites with, in some cases, the superimposition of the effects of a low-temperature prograde metamorphism. The great differences in the variation patterns of Ni/Fe and Ni/Co from one chondrite to another even within the same group and petrological type, as well as the differences in the average Ni and Co contents of the metal phases, suggest a convenient method for "finger-printing" meteorites.

Keywords: meteorite, LL type, brecciated chondrite, Innisfree, Alberta, electron-microprobe analyses.

Sommaire

La météorite d'Innisfree, de type LL, est une chondrite bréchifiée et probablement génomicte. Plagioclase, orthopyroxène, clinopyroxène, olivine, chromite, ilménite, merrillite (= whitlockite), apatite, troïlite, kamacite, taenite et chondrules dévitrifiées ont été analysés à la microsonde électronique. Les rapports Ni/Fe et Ni/Co varient très fortement dans les phases métalliques; ces variations contrastent avec celles qui caractérisent plusieurs autres chondrites et suggèrent une évolution autométamorphique pour l'équilibration des météorites chondritiques, avec, dans certains cas, superposition d'un métamorphisme prograde de basse température. Les grandes différences dans la variation de ces rapports et dans les teneurs moyennes en Ni et Co des phases métalliques, d'une chondrite à l'autre, voire à l'intérieur d'un même groupe ou d'un type pétrologique, offrent une méthode commode d'identification des météorites.

(Traduit par la Rédaction)

Mots-clés: météorite, type LL, chondrite bréchifiée, Innisfree, Alberta, analyses à la microsonde électronique.

Introduction

The Innisfree meteorite fell near the town of that name in Alberta early in the evening of February 5, 1977 (Halliday *et al.* 1978, Revelle 1979). It is of particular interest because it is only the third recovered meteorite for which two-station photography has permitted a reliable orbital path to be calculated. This path indicates an aphelion at ~2.76 AU, well beyond Mars in the asteroid belt. As part of a thorough characterization of this meteorite, a detailed investigation of the constituent mineral phases has been carried out by electron microprobe.

General Description

In thin section it is apparent that the Innisfree meteorite has a brecciated texture and

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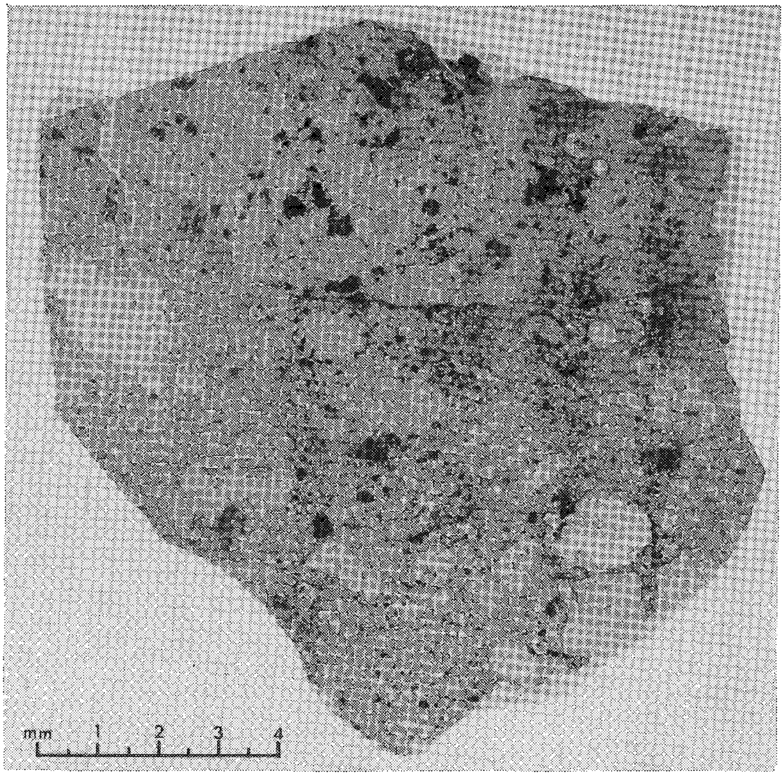

FIG. 1. The polished thin-section of Innisfree used in this work, seen in transmitted light. Note the different textures of the upper (type 6) and lower (type 5) parts, which are separated by a very narrow opaque zone exhibiting shock effects. The large white area in the upper left is a hole in the section.

that different regions of this breccia show different degrees of recrystallization, as reflected by the definition of chondrule boundaries and the grain size of the matrix (Fig. 1). Chondrules, chondrule fragments and possibly lithic fragments, which range in size from nearly 2 mm downwards, are abundant. These make up as much as 50% of the meteorite; many different varieties, including excentroradial, various other radiating and feathery types, porphyritic and barred olivine chondrules are present. Fragments are usually angular or subrounded. The matrix material ranges in size downwards from about 0.1 mm, with some of the larger grains probably breccia fragments. In the more recrystallized region, where chondrule margins are less well defined, the proportion of fine grained matrix is higher.

Olivine and pyroxene are the predominant silicates; minor plagioclase, which is well crystallized and not maskelynite, is present in the matrix. No areas of true glass were observed, although some chondrules contain turbid devitrified glass. Opaque minerals make up less than 10% of the meteorite, with troilite more abundant than metal grains. Chromite and ilmenite are rare.

The two different brecciated regions observed in the thin section of the meteorite are separated by a thin vein of opaque material. This is made up of glass that includes extremely small droplets and films of metal and sulfide. Also present in the immediate region of the boundary are occasional grains of troilite containing immiscible droplets of excess metal. Apart from minor strain effects in troilite grains, the indications of shock extend for no more than about a hundred microns from the boundary.

In view of the thin-section evidence indicating that Innisfree is a breccia, towards the end of this study it was decided to saw individual number 5 in half so that the texture could be observed more easily and over a larger area.

The cut surface (Fig. 2) confirmed the brecciated nature; irregularly shaped, dark grey fragments make up about 20% of the exposed area and are set in a lighter grey host material. In many places the dark grey material appears to have been drawn out into veins, but the contact between light and dark grey areas is generally sharp.

Thin sections of 23 LL5 and LL6 chondrites in the collections of the Department of Mineralogy, British Museum (Natural History) were examined for comparative purposes. One of these meteorites, Krähenberg, is texturally very similar to Innisfree.

Analytical Procedures

The analyses reported in Table 1 were performed using an ARL "EMX" microprobe fitted with three wavelength-dispersive spectrometers and an Ortec energy-dispersive spectrometer. Well-characterized analytical standards were used throughout. All data were obtained at an operating voltage of 15 kV and with probe currents of the order of 20 nA. For energy-dispersive analyses, collection times were 400 s at a full-spectrum counting rate of approximately 3000/s. Wavelength-dispersive results were processed using the APL program PROBEDATA (Smith & Tomlinson 1970), and energy-dispersive spectra by EDATA (Smith & Gold 1976) or EDATA2 (Smith & Gold 1979).

The determination of Ni and Fe in the metal phases was carried out by wavelength-dispersive analysis using calibration curves based on pure Ni, pure Fe and four Ni–Fe alloys of intermediate and precisely known compositions. This procedure is both accurate and extremely rapid for essentially binary systems; background and matrix corrections are automatically taken into account. However, with randomly chosen points, the excited volume will sometimes lie astride the boundary of the two phases. Although this will tend to broaden peaks in a histogrammic plot such as presented in Figure 3, the narrow widths of kamacite peaks in that plot indicate that the effect is not serious. The small amounts of Co present in the metal phases will detract only trivially from the accuracy of the technique outlined.

The Co determinations reported here were made by wavelength-dispersive analysis against a pure Co standard. Matrix corrections have not been applied because calculation showed them to be very small (0.95–0.97) throughout the entire range of metal compositions encountered.

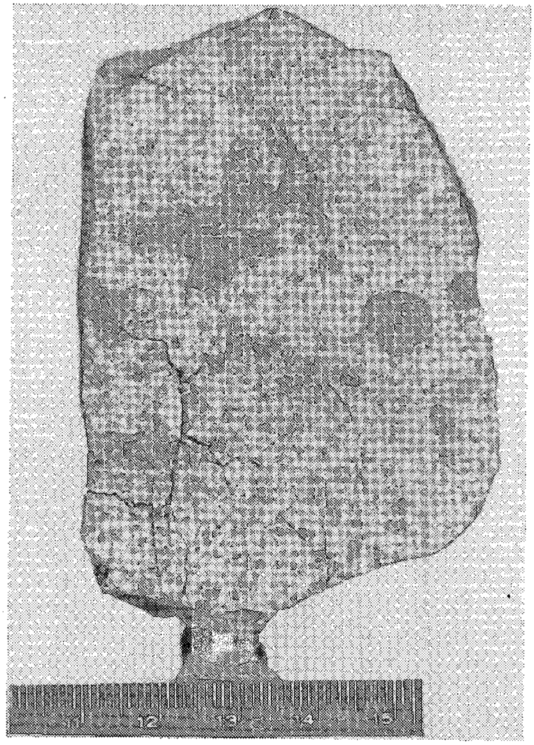

FIG. 2. The sawn surface of Innisfree individual #5 (894g) showing the brecciated texture, with dark grey areas in a lighter grey host. The scale attached to the specimen is in cm.

Results

The olivines

Eleven different grains were analyzed, and the average composition and extreme values are shown in Table 1. The mineral is rather homogeneous and has an average molecular ratio Fe/(Fe+Mg) of 0.271. This places it at the low end of the range of values associated with the LL group of chondrites (Keil & Fredrickson 1964).

The pyroxenes

Both monoclinic and orthorhombic low-Ca pyroxenes appear to be present in the meteorite. Much of the clinohypersthene shows polysynthetic twinning, suggesting that it has inverted from an earlier protopyroxene structure. In some places, there are indications of zoning in the form of narrow rims of slightly higher birefringence. Although no attempt was made to differentiate between low-Ca pyroxenes of different symmetry during analysis, the twelve

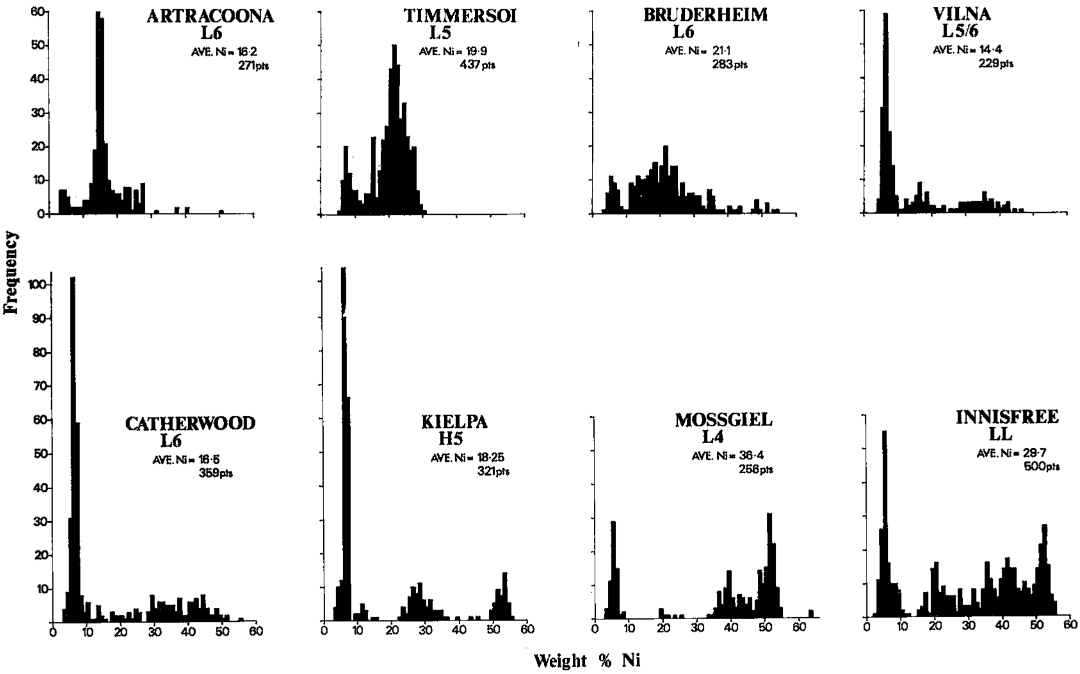

FIG. 3. Histogram plots of the wt. % Ni in the metal phase of Innisfree and some other chondrites.

grains investigated proved to be similar, at least in the molecular ratio Fe/(Fe+Mg), which averaged 0.226. This value places this mineral also at the low-Fe end of the range for LL chondrites. The minor elements Al, Ca, Ti, Cr and Mn show substantial grain-to-grain variations.

The eight high-Ca pyroxene grains analyzed exhibit more relative variation in the ratio Fe/(Fe+Mg) than is seen in the coexisting low-Ca pyroxene; the concentrations of the minor elements Na and Al also show appreciable scatter. However, the average composition of the grains analyzed is closely similar to the average for clinopyroxenes of eight group-LL chondrites quoted by van Schmus & Koffman (1967).

Plagioclase

The mineral is typically fine grained, and its composition is close to that normal for chondrites of L and LL groups (van Schmus & Ribbe 1968). Because of the fine grain-size, some alkalis may have been lost during the microprobe analysis.

The oxide minerals

Ten grains of chromite were analyzed to provide the data shown in Table 1. The mineral is rather homogeneous except for Ti, which varies substantially. When plotted on the variation diagrams of Bunch *et al.* (1967), the chromite plots in the region of other LL-group chondrites, although the plot of the FeO content of the chromite is slightly anomalous. The composition of four grains of ilmenite appears to be intermediate between the data points plotted by Snetsinger & Keil (1969) for the L and LL groups.

The phosphates

The two common phosphates of chondritic meteorites, merrillite (= whitlockite) and chlorapatite, are both present, the latter being the more abundant. The compositions of these phosphates appear to be typical of chondrites according to the data of van Schmus & Ribbe (1969) and Fuchs (1969). The merrillite contains small concentrations of F and Si, neither of which have been reported in previous analyses of terrestrial whitlockites or meteoritic merrillite. The excess of metal cations [3.16 instead of 3.00 for the ideal formula of $(Ca, Mg,Na)_3P_2O_8$] is consistent with similar excesses in other meteoritic merrillite and leads to a structural formula very close to $Ca_{18}(Mg,Fe)_2$

TABLE 1. COMPOSITIONS OF ANALYZED PHASES IN THE INNISFREE METEORITE

	OLIVINE			LOW-Ca PYROXENE			HIGH-Ca PYROXENE			PLAGIOCLASE			CHONDRULE 'GLASS'¶		
	AV	HI	LO	AV	HI	LO	AV	HI	LO	AV	HI	LO	AV	HI	LO
H															
F															
O	40.58	40.72	40.41	44.53	44.84	44.28	43.55	44.09	43.18	48.53	48.73	48.27	46.85	46.93	46.73
Na							0.33	0.53	0.19	6.25	6.72	5.91	4.77	4.87	4.59
Mg	22.02	22.54	21.56	16.75	17.22	16.14	9.94	11.22	9.38	0.07	0.12	0.00	3.86	4.21	3.60
Al	0.01	0.09	0.00	0.19	0.61	0.00	0.46	0.79	0.14	11.25	11.37	11.12	7.42	7.76	7.16
Si	18.01	18.29	17.85	25.99	26.24	25.66	25.24	25.99	24.54	31.04	31.30	30.68	28.79	28.97	28.66
P															
S													0.19	0.22	0.14
Cl													0.02	0.05	0.00
K										0.73	0.82	0.53	0.39	0.48	0.31
Ca				0.68	1.26	0.35	15.07	15.73	13.90	1.66	1.95	1.55	5.27	5.87	4.76
Ti	0.02	0.04	0.00	0.04	0.08	0.00	0.23	0.36	0.14				0.08	0.12	0.03
V															
Cr	0.09	0.55	0.00	0.21	0.56	0.03	0.50	0.64	0.41				0.18	0.19	0.16
Mn	0.35	0.38	0.31	0.36	0.40	0.26	0.13	0.18	0.11				0.03	0.05	0.00
Fe	18.84	19.46	17.94	11.23	12.21	10.00	4.55	6.06	3.45	0.46	0.66	0.27	2.09	2.39	1.72
Co															
Ni	0.08	0.12	0.00	0.01	0.05	0.00		0.05	0.00	0.01	0.05	0.00			
Zn				0.01	0.05	0.00									
Sr															
REE[1]/Zr[2]													0.06[2]	0.07	0.04
TOTAL*	100.00	-	-	100.11	-	-	100.00	-	-	100.05	-	-	99.77	-	-

	CHROMITE			ILMENITE			MERRILLITE†			APATITE†			TROILITE			KAMACITE/TAENITE†		
	AV	HI	LO	AV	HI	LO	AV	HI	LO	AV	HI	LO	AV	HI	LO	AV	HI	LO
H										0.02	0.04	0.00						
F							0.38	0.40	0.35	0.66	0.66	0.66						
O	29.92	30.17	29.82	32.18	32.33	32.15	41.42	41.42	41.41	37.40	37.81	37.40						
Na							1.91	2.02	1.85	0.23	0.23	0.23						
Mg	1.48	1.88	1.23	1.37	1.56	1.26	2.12	2.18	2.05	0.03	0.03	0.03						
Al	2.92	3.16	2.75				0.01	0.02	0.01	0.01	0.01	0.01						
Si	0.28	0.34	0.23	0.18	0.25	0.13	0.15	0.15	0.13	0.10	0.12	0.07						
P							20.04	20.05	20.04	17.84	17.89	17.80						
S													38.58	36.73	36.43			
Cl							0.01	0.02	0.01	5.00	5.67	4.33						
K																		
Ca	0.06	0.09	0.04	0.07	0.09	0.05	33.16	33.26	33.02	38.31	38.36	38.26						
Ti	1.56	2.31	1.13	31.48	31.67	31.30												
V	0.45	0.52	0.42	0.27	0.28	0.26												
Cr	37.91	38.84	36.79	0.04	0.06	0.01												
Mn	0.50	0.54	0.41	0.90	0.92	0.88	0.02	0.03	0.02	0.01	0.01	0.01						
Fe	24.60	25.58	23.67	33.49	33.65	33.32	0.70	0.79	0.64	0.34	0.52	0.15	63.38	63.57	63.25	69.80	94.00	44.30
Co													0.04	0.16	0.00	0.54	1.26	0.00
Ni				0.02	0.05	0.00										29.66	55.60	4.80
Zn	0.32																	
Sr							0.01	0.03	0.00	0.03	0.03	0.03						
REE[1]/Zr[2]							0.07[1]	0.09	0.05	0.02[1]	0.02	0.02						
TOTAL*	99.91	-	-	99.18	-	-	99.48	-	-	100.74	-	-	99.61	-	-	100.00	-	-

† Wavelength-dispersive analysis. All other results by energy-dispersive analysis. ¶ All 'glass' is turbid.

* All analyses recalculated to 100% from totals shown. § Oxygen determined by calculation.

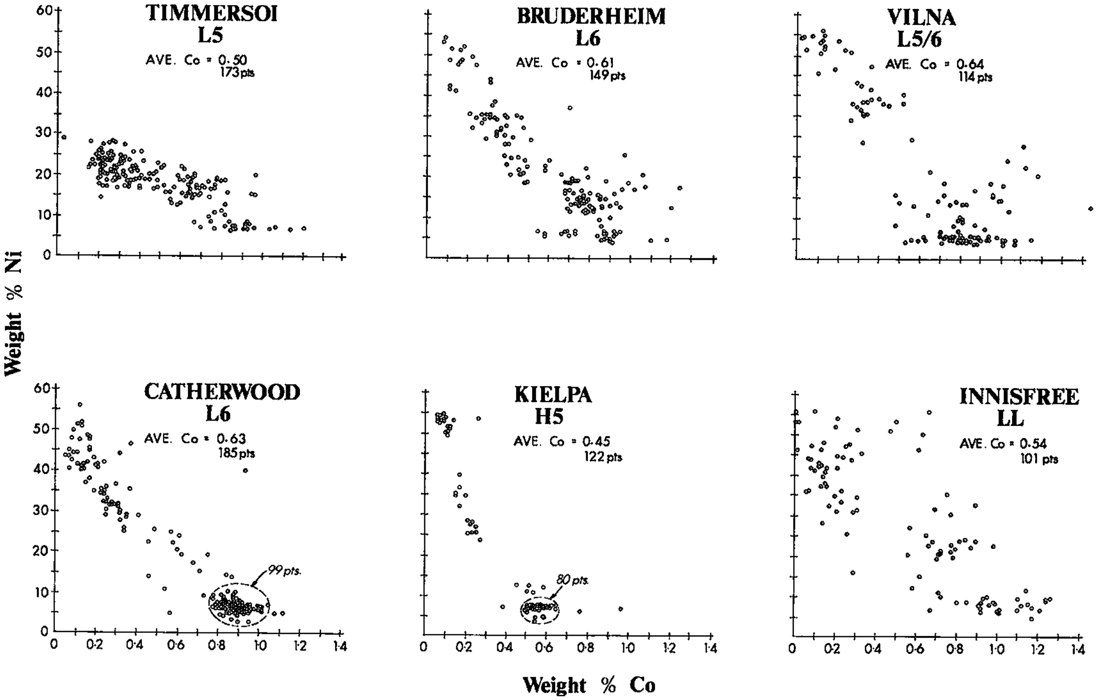

FIG. 4. Variation of Co with Ni content of the metal phases of Innisfree and five other chondrites.

$Na_2(PO_4)_{14}$, which Dowty (1977) found to be typical for the mineral occurring in chondrites. The evidence found by Dowty for structural differences between terrestrial whitlockites and meteoritic merrillites, together with the consistently different composition for the meteoritic material, suggest that the earlier name merrillite, the abandonment of which was recommended by Fuchs (1969), should now be reinstated. Accordingly, it is used throughout this paper.

Sulfide

Troilite is the only sulfide identified. It is stoichiometric within the accuracy of the analysis. Very small amounts of Ni and Cr (<0.01%) are possibly present, whereas V, Mn and Cu were sought but not found.

Kamacite and taenite

The metallic phases of the meteorite were investigated initially by making a number of randomly chosen spot-analyses for Ni and Fe. The large range of values encountered prompted a more extensive investigation involving about 500 analytical points. The results for Ni are plotted as a histogram in Figure 3. The wide range of compositions exhibited by the metal phases of the Innisfree meteorite invites comparison with other chondrites. Point analyses were therefore carried out using the same technique on a number of other chondrites available for study. Some of the results are shown in Figure 3, where it will be observed that the pattern of variation varies appreciably from one meteorite to another.

Cobalt determinations made at the same time as some of the Ni determinations on the Innisfree metal phases also indicate considerable scatter. Figure 4 shows the variation of Co as a function of the Ni content in Innisfree metals and compares this variation with that seen in five of the other chondrites used to produce Figure 3. Figures 3 and 4 also show the average Ni and Co contents of the metal phases for each of the meteorites, calculated from the determinations plotted in these figures. In the case of Innisfree, the overall average composition is Fe = 69.80, Ni = 29.66 and Co = 0.54 wt. %. These figures are unlikely to be as accurate as the averaging of 500 data points might suggest because of probable biases in selecting analytical points (*e.g.*, on the basis of grain size or away from the grain edges). However, the Ni and Fe figures are probably good to within

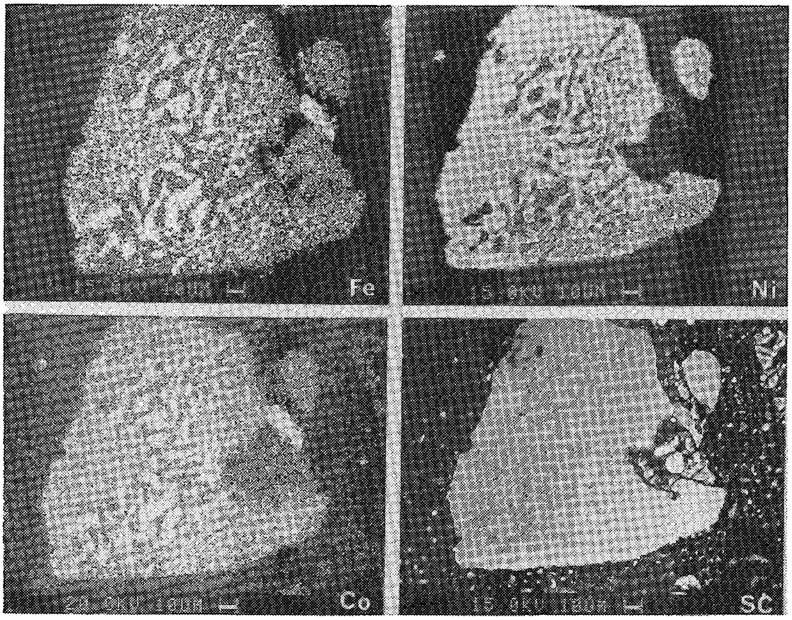

FIG. 5. Scanning images of a plessitic intergrowth of taenite and kamacite in the Innisfree meteorite. All images were obtained using an ARL "SEMQ" instrument and at the operating voltages shown beneath the photographs. Magnification is indicated by the measuring bar and the figure in microns ("UM") beside it. For the sample-current image (SC) the signal has been inverted.

2–3% relative and the Co contents probably within 5% relative. Similar, perhaps slightly poorer, accuracies should apply to the Ni figures given for the other meteorites, for which somewhat fewer determinations were made. Scanning images were obtained of grains of metal in the Innisfree meteorite. Although some grains appear homogeneous, others show complex intergrowths of Ni-rich and Fe-rich areas (Fig. 5).

DISCUSSION

The analyses presented here suggest that the olivine and orthopyroxene of the Innisfree meteorite are in equilibrium, according to the experimental findings of Medaris (1969). The presence, additionally, of high-Ca clinopyroxene allows an approximate determination of the equilibration temperature by the method outlined by van Schmus & Koffman (1967). Based on the average compositions for low-Ca and high-Ca pyroxenes, the distribution coefficient K_2 of $\sim$0.68 [$=(X_{Fe}^{cpx} \cdot X_{Mg}^{opx})/(X_{Mg}^{cpx} \cdot X_{Fe}^{opx})$] indicates an equilibration temperature of about 1020°C, a value that is appreciably higher than the temperature of 820 ± 50°C found by van Schmus & Koffman (1967) for 21 equilibrated chondrites. However, the average composition of the Innisfree low-Ca pyroxene includes data for clinohypersthene. Furthermore, using the extremes of even the rather small range of Mg and Fe values for the low-Ca pyroxenes (together with the average high-Ca pyroxene values), one obtains temperatures of either $\sim$800 or $\sim$1250°C. Moreover, the two-pyroxene thermometer, based on the partition of elements between the solids, is inapplicable if equilibrium was established in the presence of a silicate liquid phase either present during initial crystallization or produced as a result of some later reheating event. In these circumstances the significance of the temperature calculated for Innisfree remains uncertain.

Urey & Mayeda (1959) drew attention to the great variety of textures and composition present amongst metal phases in chondritic meteorites. Wood (1967) showed how these might be interpreted in terms of the Ni–Fe phase diagram and individual histories involving different rates of cooling with, in some cases, the superimposition of effects due to a later reheating event. More recently Bevan & Axon (1980) have described the metal phases in the unequilibrated H3 chondrite, Tieschitz. The polycrystalline intergrowths of kamecite, taenite and troilite and the multizoned taenite led them to postulate the preservation of relict primary solidification textures during rapid cooling to temperatures of about 700°C, whereas α/γ interface compositions of ~5% Ni and ~50% Ni indicated an equilibrium established at about 350°C. But in equilibrated type-5 and -6 chondrites, in which silicates and oxides invariably show very restricted compositional ranges within a given meteorite, the great ranges and varieties of Ni/Fe values encountered in the metal phases are remarkable. It is difficult to conceive of a single metamorphic heating-and-cooling cycle producing these Ni/Fe values (or, for that matter, the sort of textural features illustrated in Figure 5), unless perhaps it was a cycle during which the rate of change of temperature fluctuated greatly. One could more easily envisage the slow cooling of chondritic material from the temperature at which it solidified, producing a high degree of homogeneity in the silicates and oxides with equilibration reactions commonly continuing down to temperatures of about 800°C. Then, as the temperature continuel to fall, inhomogeneity in the metal grains would be produced by exsolution, the process proceeding towards completion to a different extent in grains of different size. Such zoned and inhomogeneous grains, possibly further modified during the accidental processes of agglomeration, shock and brecciation, would each behave somewhat differently according to their size, the phases present and their compositions, during any subsequent, prolonged, low-temperature (300–400°C) metamorphism produced by burial in a "parent" body.

Afiattalab & Wasson (1980) have recently confirmed the findings of Sears & Axon (1976) that the Co contents of kamacites in ordinary chondrites show nonoverlapping ranges. These ranges are: H group 0.33–0.44%, L group 0.67–0.82%, and LL group 1.5–11%. The Co values reported here for six other ordinary chondrites do not support such a neat and straightforward pattern of variation. Thus, none of the Innisfree kamacite has a high enough Co content to place it within the range 1.5–11%, whereas values for Catherwood kamacite give an average above the maximum of 0.82% for the L group, and the average Kielpa kamacite has a Co value of about 0.58%. Afiattalab & Wasson (1980) suggest that there is a spectral interference between a low-intensity "forbidden" line of Fe ($K\beta_o$) and the Co analytical line, and that this would result in Co values in kamacite that are consistent high (by ~0.17%) unless a correction were applied. If this were indeed the case, the average values for Catherwood and Kielpa would lie within or close to the ranges that Afiattalab & Wasson propose, but Innisfree would be even more deficient in Co. Furthermore, it is difficult to see why a significant number of the Co data points for taenite (with about 50% Fe) should show concentrations $\leqslant$ 0.05% (*e.g.*, Vilna and Innisfree). In this regard, it is perhaps significant that Afiattalab & Wasson obtained negative values for the Co content of taenite grains in some H-group chondrites when they applied their correction. Long & Hendry (J.V.P. Long, priv. comm., 1980) investigated the satellite line in question in some detail and found that its intensity as measured with a "Geoscan" microprobe at 20 kV is less than 0.0005 x Fe $K\alpha$ peak intensity (or less than about 20% of background). Furthermore, the satellite peak intensity is displaced by about 0.2° 2θ from the Co $K\alpha$ peak so that its contribution to the observed Co counts would be even less than 0.0005 x Fe $K\alpha$ intensity. It seems possible that in measuring the intensity of this satellite line, Afiattalab & Wasson failed to make the appropriate correction for background, a correction that is indeed difficult in the presence of the intense adjacent Fe $K\beta_{1,3}$ peak.

There is a great deal of scatter in the Co values of the metal grains of some of the chondrites investigated here. The extent of this scatter seems to be related roughly to the spread in taenite compositions. Thus Innisfree, which shows the greatest range of taenite compositions and the highest proportion of Ni-rich taenite, also shows the most scatter in the Co values. At the other extreme, Kielpa shows a scatter of Co values about the mean for particular kamacite or taenite compositions that is only slightly greater than the probable analytical error.

The apparent absence of very high Co values in metal grains from Innisfree could reflect the fact that in this work no attempt was made to measure Co contents at the extreme edges of grains because of the many errors that can be introduced in such analyses. It is possible, therefore, that high values do exist: if so they will tend to increase the average Co content of Innisfree kamacite. No such situations were observed, however, in the scanning images that were examined (see, for example, Fig. 5).

Conclusions

The evidence of brecciation and the presence of two different petrological types belonging to the same (LL) group within one thin section suggest that Innisfree is possibly a genomict chondrite according to the definition of Wasson (1974), although this conclusion might be altered in the light of further microscopy and perhaps analytical work on other parts of the breccia. The textures present in the least recrystallized regions examined in thin section (the lower part in Fig. 1) are characteristic of petrological type 5. The average compositions of the silicates and oxides in both type-5 and type-6 parts are very similar; the olivine, pyroxenes and oxides all indicate that Innisfree can best be classified as a member of the LL group, although in nearly every respect it lies at the extreme end of the range of parameters that can be used to characterize that group, in the direction of L-group parameters.

Additional studies that integrate detailed metallography, electron-scanning imagery and accurate micro-analytical work on a much more comprehensive suite of chondrites will be required if the role and importance of each factor possibly involved in producing the patterns of variation of the Ni, Fe and Co concentrations are to be evaluated. One would not wish to speculate in detail on the implications of these variations until such additional investigations were carried out. However, it would seem that they tend to favor an origin for chondrites involving equilibration principally during initial cooling with, in some instances, minor low-temperature burial metamorphism producing additional effects on the relatively sensitive metals. This is quite consistent with the suggestion of Scott (1979) that the metamorphic grades of chondrites may have been produced in kilometre-sized planetesimals that subsequently accreted into 10–100 km bodies, where mild metamorphism and slow cooling occurred.

The large numbers of analyses used to produce the histograms of Figure 3 are relatively easily obtained, especially with the fully automated microprobes now available. The wide range of different patterns already observed suggests that the determination of the Ni–Fe distribution patterns and also the average Ni and Co contents of metal phases may prove very useful in finger-printing individual meteorites. Such a technique could be applied to establishing affinities amongst large groups of meteorites such as those recently recovered from Antarctic blue ice (Nagata 1976, Cassidy *et al.* 1977). It may also be of use in solving problems concerning the identity of certain specimens collected from different localities or perhaps mislabeled specimens in meteorite collections.

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