Distribution of Copper, Cobalt and Nickel in Ores and Host-Rocks, Ingladhal, Karnataka, India

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Stratiform quartz-sulphide lodes in Ingladhal occur in a typical Precambrian green-stone-belt environment comprising metabasalts, tuff, chert and cherty iron-sulphide formation. Unusually high cobalt contents of metavolcanics and of sulphide minerals in orebodies suggest a consanguinity between ores and rocks. 90% of total nickel, 70% of total cobalt but only 30% of total copper in rocks occur in silicate phases and thus indicate an early separation of copper from cobalt and nickel. Unusually high non-sulphide copper in barren bedded cherts implies availability of Cu-rich solution prior to their lithification. Pyrite in sediments, in volcanics, and in orebodies is characterized by a distinctive pattern of Co-Ni distribution in each case. Partitioning of Co and Ni between coexisting sulphide pairs is complex, but gross equilibrium is indicated. Very high trace metal content of orebody pyrite sharply contrasts with very low such values in pyrite from adjacent sediments and points to a higher temperature of formation of orebodies.

SYMBOLS

Py - Pyrite
Po - Pyrrhotite
Cpy - Chalcopyrite
i, j, etc - different elements
α, β, etc - mineral phases

X_i^{α}; X_{Ni}^{Py} - concentration of element i in phase α; concentration of nickel in pyrite

$(i/j)_{\alpha}$; $(Co/Ni)_{Py}$ - ratio of concentrations of elements i and j in phase α; ratio of Co:Ni in pyrite

$R_i^{\alpha-\beta}$; R_{Co}^{Py-Po} -distribution of an element i between the phases α and β as determined by X_i^{α}/X_i^{β}; ratio of cobalt in pyrite to cobalt in pyrrhotite.

INTRODUCTION

Use of trace element data of ores and host-rocks, in order to characterize the environment of ore deposition and to establish the genetic nexus between ores and the supposed parent rocks, is often beset with severe limitations. The initial high hopes raised by earlier studies (e. g. Auger, 1941; Davidson, 1962; Hawley, 1952; Hawley and Nicol, 1961; Hegemann, 1943) have been belied, mainly through later careful investigations which have revealed numerous unforeseen complexities. Nevertheless, such data, when used in conjunction with relevant geologic information, have proved rewarding (e. g. Cambel and Jarkovsky, 1967, 1969; Go-

vett and Pantazis, 1971; Loftus-Hills and Solomon, 1967). Mercer (1976), in his review article, has emphasized the limitations and potentialities of the uses of trace element data of metal deposits in sedimentary rocks.

During the course of an investigation of the Ingladhal (14°8' 28" - 14°12' 0"N; 76°26'8" - 76°28' 16"E) copper deposit (Philip, 1971, 1977; Bagchi, Mookherjee and Philip, 1970) the patterns of distribution of cobalt nickel and copper in the country rocks and of cobalt and nickel in individual sulphide minerals were examined. The present report incorporates results of the geochemical investigation. A brief summary of the geological setting, mainly based on Philip's (1971, 1977) report, is also incorporated.

Trace elements were determined by a model 303 Perkin Elmer atomic absorption spectrophotometer. Rock samples were first microscopically examined to ensure that these were "unmineralised" and powdered samples were subjected to repeated digestion in HF-Perchloric acid. The sulphide minerals were separated by a Franz Isodynamic Separator, heavy liquid and, when necessary, by handpicking under the binocular microscope until > 98% purity was attained. Sulphide samples were digested in Conc HNO_3 and perchloric acid to obtain the solution. Blanks were prepared in the same fashion.

GEOLOGIC SETTING

The Ingladhal ore deposit, located 6 km south of Chitaldurg (also known as Chitradurga) township in Karnataka, occurs within the Archaean greenstone belt of southern India. As such, many of the distinctive features of stratabound sulphide deposits in Precambrian volcanic milieu (Gilmour, 1971; Goodwin and Ridler, 1970; Sangster and Scott, 1976) are characteristically present.

The area consists predominantly of a differentiated suite of lava-flows and tuffaceous rocks, with varying amounts of conglomerates cherts and sulphide

facies of iron-formation. The lava-flows, often with well-preserved pillows, are mostly basaltic, - more felsic varieties such as andesite, trachyte and keratophyres constituting about 10% of the total volume of volcanics. Philip (1977) has described nine varieties of welded tuffs, which include fine laminated ash-flows, crystal tuff, reworked tuff etc, from this region. Sedimentary rocks are conformable with the volcanics and generally occupy higher stratigraphic horizons. Massive chert bands are interlayered with the tuffaceous rocks. A banded pyrite-pyrrhotite-chert rhythmite constitutes a marker horizon for a strike length of more than 10 km in the immediate vicinity of the ore deposit and represents the sulphide facies of iron-formation. Interlayering of volcanics with sediments, presence of acid volcanic differentiates (though quantitatively insignificant) and rarity of coarse pyroclastics point to a relatively quiescent, waning phase of volcanism. Younger dolerite dikes cut through the metavolcanics and metasediments.

Grade of regional metamorphism is low to moderately low; tuffaceous rocks are represented by fine-grained chlorite schists with well preserved primary laminations, basalts by chlorite/actinolite schists and epidote-amphibolites, and pyritic cherty formations by pyrite-pyrrhotite-chert rhythmite.

Deformation of these rocks were predominantly through compression in the E-W direction, - structural elements having evolved in two successive stages. Two distinct fold styles have been recorded. One isoclinal, with sharp crests and varying trends and amounts of plunge; the other, an open fold system with steep plunge and near-constant trend. Refolding of the (earlier) isoclinal folds is represented by twisted minor-fold axes and bent axial plane traces of earlier folds. On a regional scale, the area forms part of a major southerly-plunging anticline, as shown by structural analysis (Philip, 1971). Copper mineralization occurs as stratiform quartz-sulphide lodes within what has been variously described as "schistose rocks" (Viswanathaiah et al.,

1971) or "highly schisted volcanic flows" (Radhakrishna, 1974). Careful petrographic examination by Philip (1977) reveals that the immediate host rock is a variety of layered "cleaved tuff" occurring interbanded with variolitic traps and banded pyrite-pyrrhotite rhythmite. Main quartz-sulphide lodes are conformable with the lamination in tuff while "cross-veins" (Viswanathaiah et al., 1971), athwart to primary banding, occupy S-planes developed during the two phases of folding. Mineralogy of the ore is rather simple. Pyrite pyrrhotite and chalcopyrite constitute about 95% of the sulphide minerals. Minor amounts of sphalerite galena and boulangerite are reported (Pathan and Naganna, 1970; Borreswarrao and Acharyulu, 1970). The ore reserve, according to Radhakrishna (1974) one mill. t. of 1.4 % Cu.

PREVIOUS WORKS

Geology of the area and aspects of mineralisation have been described by Radhakrishna (1967, 1974) and Viswanathaiah et al. (1971). Borreswarrao and Acharyulu (1970) have described the mineralogy and paragenesis of the sulphide ore minerals. Naqvi and Hussain (1973) have discussed the geochemistry of the metavolcanics. Philip (1977) has described the petrographic varieties of tuffs present in the area and proposed their mode of origin. Hegde and Aswathanarayana (1977) have discussed the ore elements in the metavolcanics of the area and the possibility of using the data for ore prospecting. Bagchi, Mookherjee and Philip (1970) briefly discussed the geology and metallogenesis of the area.

TRACE METALS IN THE COUNTRY ROCK

Results

Thirty seven unmineralised samples (three cherts, eight tuffs, fifteen lava-flows and basic intrusives and eleven chlorite/amphibole schists) of the country rock were analysed for copper cobalt and nickel. The results are shown in Table 1.

The three chert samples, with very low Co and Ni in nearly equal proportions and very high copper content, stand apart as a group from the rest of the analyses. One of the samples is from sulphide-free portion of the pyrite-pyrrhotite-chert band, and the other two are from unmineralised thick chert bands (interlayered within the tuffs) that do not show a single sulphide grain under the microscope.

The rest of the metavolcanics, comprising various types of tuffs, traps, younger dolerites and schists (but predominantly basaltic), gives an average assay of 146 ppm Ni, 66 ppm Co and 67 ppm Cu. Its correspondence with the data of Naqvi and Hussain (1973) is rather striking (124 ppm Ni, 66 ppm Co, and 68 ppm Cu), even though they covered a larger area than that of the present investigation.

Among the volcanics, the tuffs are distinguished by (a) a much lower nickel content (average 78 ppm) than the general average (146 ppm Ni), and (b) a much higher Co/Ni ratio which exceeds 1.0 in four out of eight samples.

The lava-flows and their metamorphosed equivalents together with the younger dolerites give an average assay of 168 ppm Ni, 67 ppm Co, and 72 ppm Cu. But for some excess in nickel, this average is virtually similar to the average values when the tuffs are also included.

Discussions

Hegde and Aswathanarayana (1977) have studied the leachable (i.e. sulphide fraction, released by H_2O_2 - ascorbic acid mixture) trace metal content of the metavolcanics around Ingladhal. Their average assay (Ni-18 ppm, Co-12 ppm, Cu-46 ppm), when compared with those of the present study and Naqvi and Hussain's (1973), immediately suggests that about 90% of Ni, 70% of Co, but only 30% of

Table 1. Cobalt nickel and copper content (ppm) of unmineralized rocks from Ingladhal

Sl. No.	Rock Type	Sp No.	Ni	Co	Cu	Co/Ni
A. Stratiform chert bands						
1.	Chert	R 21	10	< 10	430	
2.	Chert-Fe-sulphide rhythmite (sulphide free portion)	R 94	10	< 10	450	
3.	Chert	R 404	35	20	460	0. 6
		Average	18	-	447	
B. Tuffs of diverse texture						
1.	Tuff	R 1	125	60	80	0. 48
2.	Tuff	R 45	125	45	85	0. 36
3.	Tuff	R 272	50	50	60	1. 00
4.	Tuff	R 296	20	35	10	1. 75
5.	Tuff	R 586	115	70	50	0. 61
6.	Tuff	R 309	90	54	65	0. 60
7.	Tuff	R 134	65	70	80	1. 07
8.	Tuff	R 266	30	60	15	2. 0
		Average	78	55	56	0. 71
C. Metavolcanics and younger basic intrusives						
1.	Metabasalt	R 511	85	65	120	0. 76
2.	Metabasalt	R 585	125	60	45	0. 48
3.	Metabasalt	R 507	100	80	15	0. 80
4.	Metabasalt	R 314	450	60	70	0. 13
5.	Metabasalt	R 336	350	80	65	0. 23
6.	Variotitic "trap"	R 541	110	60	60	0. 55
7.	Variotitic "trap"	R 61	120	55	70	0. 46

Table 1. continued

Sl. No.	Rock Type	Sp. No.	Ni	Co	Cu	Co/Ni
8.	Variotitic "trap"	R 126	110	50	70	0.45
9.	Trap	R 38	300	70	70	0.23
10.	Trap	R 305	140	60	50	0.42
11.	Younger olivine norite	R 501	300	105	70	0.35
12.	Younger dolerite	R 367	130	65	30	0.50
13.	Younger dolerite	R 194	120	90	100	0.75
14.	Younger dolerite	R 159	220	70	100	0.32
15.	Olivine norite	R 499	530	65	65	0.12

D. Hornblende, tremolite and chlorite schists

Sl. No.	Rock Type	Sp. No.	Ni	Co	Cu	Co/Ni
1.	Coarse tremolitic trap	R 324	90	70	10	0.78
2.	Coarse tremolitic trap	R 312	120	80	80	0.67
3.	Hornblende schist	R 36	100	55	140	0.55
4.	Hornblende schist	R 37	85	55	70	0.65
5.	Hornblende schist	R 199	100	55	10	0.55
6.	Hornblende schist	R 207	185	65	95	0.35
7.	Hornblende schist	R 205	80	65	95	0.81
8.	Hornblende schist	R 203	80	80	75	1.0
9.	Hornblende schist	R 583	125	60	45	0.48
10.	Hornblende schist	R 523	80	105	170	1.16
11.	Chlorite schist	R 328	120	75	70	0.63
	Average (B, C & D)	-	146	66	67	
	Average (C & D)	-	168	67	72	

copper in the Ingladhal volcanic suite occur as non-sulphide, presumably locked up within the silicate phase[1]. This points to a distinct geochemical differentiation of copper from Co and Ni. The most plausible explanation is that copper, unlike cobalt and nickel, was enriched in the re-

[1] Note added in the revised manuscript. Subsequent spectrophotometric determinations of H_2O_2 - ascorbic acid leachable Cu, Co and Ni in 26 metavolcanics, 8 tuffs and 8 bedded chert samples have yielded the following values (all in ppm).

Table 2. Cobalt, nickel and copper content of Ingladhal metavolcanic suite, compared with those in basic rocks from other areas

|	Rock type, location and source of data	Cobalt (ppm) Range average		Nickel (ppm) Range average		Copper (ppm) Range average	
1a.	Ingladhal (Chitaldrug) metavolcanics excluding tuffs (present study)	50-105	67	80-530	168	10-170	71
1b.	Ingladhal (Chitaldrug) metavolcanics including tuffs (present study)	35-105	66	10-530	146	10-170	67
2.	Chitaldrug schist belt metavolcanics - Naqvi and Hussain, 1973	36-101	66	51-208	124	7-240	68
3.	Quartz-normative tholeiite - Prinz, 1967	7-65	38	0-350	76	0-300	135
4.	Olivine-normative tholeiite - Prinz, 1967	20-75	43	0-350	126	3-200	75
5.	Oceanic tholeiite - Engel et al., 1965	-	32	-	97	-	77
6a.	Troodos pillow lava (altered basalt)	-	54	-	127	-	89
6b.	Troodos pillow lava (basalt)	-	46	-	152	-	69
6c.	Troodos pillow lava (" very basic basalt")	-	54	-	207	-	76
6d.	Troodos pillow lava (olivine basalt) - Govett and Pantazis, 1971	-	59	-	348	-	74
7.	Basaltic rocks - Turekian and Wedepohl, 1961	-	48	-	130	-	87
8.	Basic rocks - Vinogradov, 1962	-	45	-	160	-	100
9a.	Depleted Archaean tholeiite (DAT)	-	60	-	225	-	110
9b.	Enriched Archaen tholeiite (EAT)- Condie, 1976	-	50	-	100	-	110
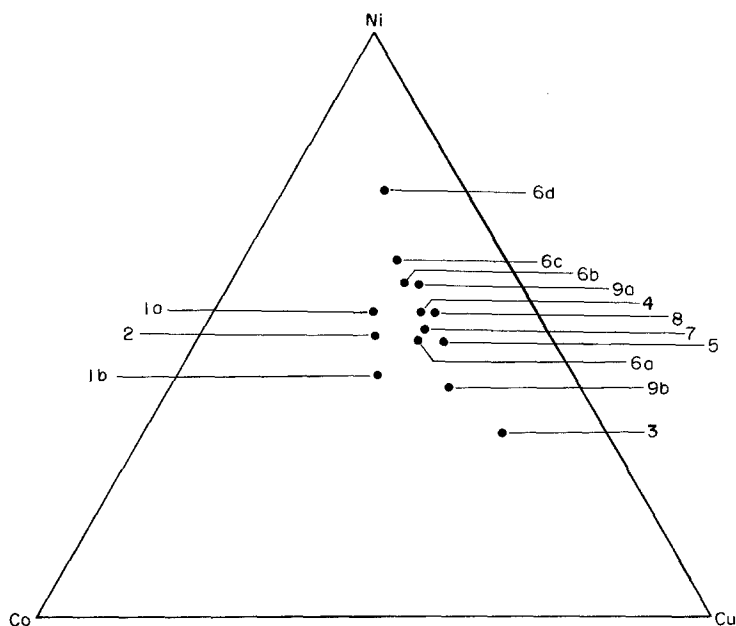
Fig. 1. Relative proportions of Cu, Ni and Co in Ingladhal metavolcanics and in similar rocks from other areas (numbers same as in Table 2)

Rock type	Copper		Nickel		Cobalt	
	mean	range	mean	range	mean	range
Meta-volcanics	52	5-110	16	6-35	18	10-60
Tuff	40	5-100	8	2-30	17	8-55
Bedded chert	15	0- 40	tr	0- 5	tr	0- 5

sidual aqueous fluid (and later precipitated as sulphides).

Trace element concentration pattern in the cherts seems to bear out the contention. Cherts are believed to have originated by direct precipitation of silica gel in places where lava flows enter the sea or where hot springs associated with volcanic activity supply water to the sea. Such cherts are well-known for their negligible amounts of trace metals (Krauskopf, 1955). But availability of Cu-rich aqueous fluids (of volcanic affiliation) before or during diagenesis, could drastically change the picture and cause copper enrichment of the cherts through adsorption by silica gel. Unusually high non-sulphide copper content and very low tenors of cobalt and nickel of the Ingladhal cherts elegantly matches the postulated situation.

It is an empirical observation repeatedly made by several workers in the Canadian shield (Goodwin and Ridler, 1970;

Ridler, 1971a; Hutchinson et al. , 1971) that there exists a spatial correlation between "exhalites" (cherts and different facies of iron formations) and volcanogenic base metal sulphide accumulation. The depositional environment most favourable for base metal sulphide accumulation is the one where coexisting iron-formation belongs to the sulphide facies, implying a shelf-to-basin transition of environment. As Ridler (1971b) summarizes it: "wherever a volcanic complex is within the sulphide or sulphidic carbonate facies and exhaling base or precious metals, significant mineralization may occur". The high copper content of cherts (and the cherty portions of the sulphide facies iron-formation) and the close spatial association of base-metal mineralisation and sulphide facies of iron formation at Ingladhal assume added significance in this context. Sangster and Scott (1976) quote Lickus (1965) and Sakrison (1966) who have shown that "the chemistry of a cherty tuffaceous layer, stratigraphically equivalent to the Vauze and Lake Dufault ore bodies in the Noranda area, significantly changes its character as the ore zones are approached".

The lower nickel content (and the high Co/Ni ratio) of the tuffs relative to the volcanic group as a whole, is a reflection of their volcaniclastic origin and is related to the greater ease of decomposition of ferromagnesian minerals (- the principal repository of nickel -) in these fine-grained rocks under subaqueous weathering conditions. These tuffaceous rocks, for which pyroclastic ash-flow, froth-flow and reworking mechanisms have been proposed (Philip, 1977), are predominantly basic andesite in composition. In the virtual absence of data on trace element content of tuffaceous rocks, a comparison of the trace elements of the Ingladhal tuffs with those of andesites is given below (data from Taylor, 1968):

The data clearly show that compared to even the most basic variety of andesites, the Ingladhal suite of tuffaceous rocks is appreciably richer in cobalt and nickel.

Table 2 presents a comparison of the trace metal content of the Ingladhal suite of volcanics with that of various types of basaltic rocks, as computed by several workers. Naqvi and Hussain (1973) have drawn attention to the fact that the average values of Co, Ni, Cu (and V) of the chital-drug volcanic suite are very similar to those reported from olivine-normative tholeiite. However, a degree of caution is warranted in such correlations, as Condie (1976) has demonstrated that all Archaean volcanic rocks, regardless of composition, are enriched in transition metals, compared to modern varieties. Similarity of the Ingladhal volcanics (excluding the tuffs) with the Troodos pillow lava basalt is rather striking. Compared to Archaean tholeiites (Condie, 1976) the Ingladhal suite (like the Troodos pillow-lava suite) is somewhat richer in cobalt and poorer in copper. Govett and Pantazis (1971) have concluded from their study that copper mineralization in the Troodos pillow lava series occurs characteristically in areas of low copper and high cobalt. Same appears to be the case at Ingladhal. Fig. 1 shows the relative proportions of Cu, Co and Ni in all the rocks mentioned in Table 2. The Ingladhal sample data plot away from the rest, towards the cobalt corner.

Four genetically-significant points, thus, emerge from the study of trace element distribution pattern in the Ingladhal metavolcanics.

a) The Ingladhal metavolcanics are richer in cobalt (with higher Co/Ni ratio) and poorer in copper than their counterparts elsewhere. This may reflect a regional characteristic feature.

Element	Ingladhal tuff	Low-Si andesite	Low-K andesite	Andesite	High-K andesite
Ni (ppm)	78	28	15	18	3
Co (ppm)	55	28	20	24	13

Table 3. Cobalt and nickel content (ppm) of sulphide minerals from Ingladhal

A. Pyrite

(i) In quartz-sulphide lodes

Sl. No.	Sp. No.	Ni	Co	Co/Ni
1	R 470	680	4060	5.97
2	R 536	385	3480	9.04
3	R 535	1700	4600	2.71
4	R 469	1150	2250	1.96
5	R 468	470	4200	8.94
6	R 442	630	3000	4.76
7	R 472	840	4650	5.54
8	R 443	645	3440	5.33
9	R 474	300	1000	3.33
10	R 1-A	410	930	2.27
11	R 43-A	680	13900	20.44
12	R 473	360	1000	2.77
13	R 531	570	15300	26.84
14	R 384	590	4160	7.1
15	R 572	165	1410	8.55
16	R 544	740	12700	17.16
17	R 102	545	1920	3.52
18	R 5-A	360	2400	6.66
19	R 74	150	1280	8.63
20	R 580	500	1370	2.74
21	R 463	470	480	1.02
Average		587	4168	7.4
Range		150-1700	480-15300	1.02-26.8

b) While nickel and cobalt in the metavolcanics occur predominantly in the silicates, copper occurs mostly as sulphide, thus implying a geochemical differentiation of copper from cobalt and nickel at a very early stage.

c) The unusually high (non-sulphide) copper content of the cherts, normally well-known for their very low trace metal content, is suggestive of local availability of copper-rich solution before or during diagenesis.

d) Very high copper content ($\approx$ 500 ppm) of unmineralized chert bands, proximity of sulphide facies of iron-formation, and lower-than-usual copper content but higher-

Table 3. continued

(ii) In cherty iron-sulphide formation

Sl. No.	Sp. No.	Ni	Co	Co/Ni
1	R 240	85	90	1.1
2	R 244	80	105	1.3
3	R 91	135	200	1.5
4	R 21	180	370	2.1
5	R 542	70	100	1.4
6	R 85	110	135	1.2
7	R 94	60	260	4.3
Average		103	180	1.75
Range		60-180	90-370	1.1-4.3

(iii) Disseminations and veinlets in tuffs and traps

Sl. No.	Sp. No.	Ni	Co	Co/Ni
1	R 284	300	800	2.7
2	R 388	240	380	1.6
3	R 110	360	1500	4.2
4	R 99	560	4940	8.8
5	R 77	380	500	1.3
6	R 475	450	1640	3.6
7	R 448	415	280	0.7
8	R 477	525	910	1.7
Average		404	1369	3.4
Range		240-560	280-4940	0.7-8.8

Table 3. continued

B. Chalcopyrite

(i) In quartz-sulphide lodes

S1. No.	Sp. No.	Ni	Co	Co/Ni
1	R 43-A	65	550	8.5
2	R 470	110	670	6.1
3	R 573	105	460	4.4
4	R 543	60	520	10.4
5	R 544	65	460	7.1
6	R 5-A	70	260	3.7
7	R 94	70	160	2.3
8	R 472	130	750	5.8
Average		85	524	6.2

(ii) Disseminations in basic lava flows

S1. No.	Sp. No.	Ni	Co	Co/Ni
1	R 475	220	480	2.2
Average		98.3	478.9	4.9

C. Pyrrhotite

In quartz-sulphide lodes

S1. No.	Sp. No.	Ni	Co	Co/Ni
1	R 74	320	400	1.3
2	R 94	200	200	1.0
3	R 468	430	3920	9.1
4	R 472	410	4060	9.9
5	R 474	330	750	2.3
6	R 544	230	3850	16.6
Average		320	2197	6.9

than-usual cobalt content of the basic me-
tavolcanics appear to be the four favour-
able attributes of the mineralized section
of the Chitaldrug schist belt. Future ex-
ploration programme within the belt
should take cognizance of these factors.

COBALT AND NICKEL IN ORE MINERALS: RESULTS

Distribution of cobalt and nickel has been
determined in 36 pyrite, 9 chalcopyrite
and 6 pyrrhotite samples. These are
shown in Table 3. Only such samples,
where chalcopyrite and/or pyrrhotite
showed textural equilibrium with unzoned
pyrite, were chosen for analysis, with a
view to determining the partition coeffi-
cients of cobalt and nickel between co-
existing sulphide phases.

Pyrite

35 out of 36 samples show $Co/Ni > 1$, the
highest value being 27:1. The only sample
(448) with Co/Ni ratio less than one (0.67)
shows sulphide disseminations confined
within fragmental ejecta embedded in la-
minated tuff.

Pyrite samples are grouped into three
categories, based on their association
and modes of occurrence: (a) those in
stratabound quartz-sulphide lodes, (b)
those in pyrite-pyrrhotite-chert rhythmite,
and (c) those as veinlets and dissemina-
tions in the metavolcanics. It is signifi-
cant that the patterns of distribution of
cobalt and nickel in these three groups
are also quite distinctive.

Pyrite from the quartz-sulphide lodes
shows maximum tenors of both cobalt
(15,300 ppm) and nickel (1700 ppm) as
well as the highest Co/Ni ratio (27:1).
The average of 21 analyses is as follows:
Ni - 587 ppm, Co - 4168 ppm; Co/Ni -
7.4:1.

Pyrite from the pyrite-pyrrhotite-
chert rhythmites, like their sulphide-
free portions, are characterised by very
low tenors of cobalt (average 180 ppm)
and nickel (average 103 ppm) and relative-
ly low Co/Ni ratio (1.7:1).

The third group - pyrite veinlets and
disseminations in metavolcanics - occu-

pies a somewhat intermediate position in
terms of the trace metal content. The
average values for eight analyses are:
Co - 1369 ppm, Ni - 404 ppm, Co/Ni -
3.4:1. Further, the group shows a very
narrow range of nickel concentration
(240-560 ppm) while cobalt concentration
ranges between 280-5000 ppm. Figure 2
shows X_{Co}^{Py} Vs X_{Ni}^{Py} plot of all analysed
pyrite samples. The diagram shows a
general tendency towards covariance of
the two elements. This is rather surpris-
ing, in view of the diverse origin and
modes of occurrence of pyrite. The field
for stratabound pyrite-pyrrhotite-chert
rhythmite can be sharply delineated; the
other two fields overlap, though the quartz-
sulphide lode samples mostly plot on the
high-cobalt side.

Pyrrhotite

Six pyrrhotite samples, coexisting with
pyrite, were analysed. A narrow range of
nickel values (200-410 ppm), a much
larger range of cobalt values (200-4060
ppm) and a wide range of Co/Ni ratios
(1-16.8) are registered. Average values
are: Ni - 320 ppm, Co - 2197 ppm, Co/Ni -
6.9:1. Samples 94 and 74 which give the
lowest Co/Ni ratio, are from the intersec-
tions of "cross-veins" with a pyrite-
pyrrhotite-chert band. The low tenors of
cobalt and nickel in these samples are
probably due to intermixing of pyrrhotites
of two different types.

Plot of X_{Ni}^{Po} Vs X_{Co}^{Po} gives a scattered
diagram (Fig. 3).

Chalcopyrite

Eight out of nine samples are from quartz-
sulphide lodes and one (475) is from pyro-
clasts embedded in tuff. Cobalt concen-
tration ranges between 160-750 ppm and
nickel concentration between 50-130 ppm.
Significantly, sample 475 records lowest
Co/Ni ratio, emphasizing its different
origin.

Plot of X_{Ni}^{Cpy} Vs X_{Co}^{Cpy} gives a relative-
ly smooth linear variation diagram (Fig.
4) particularly if sample 475 is ignored.

⊙ Quartz - sulphide lode

⊕ Pyrite - chert rhythmite
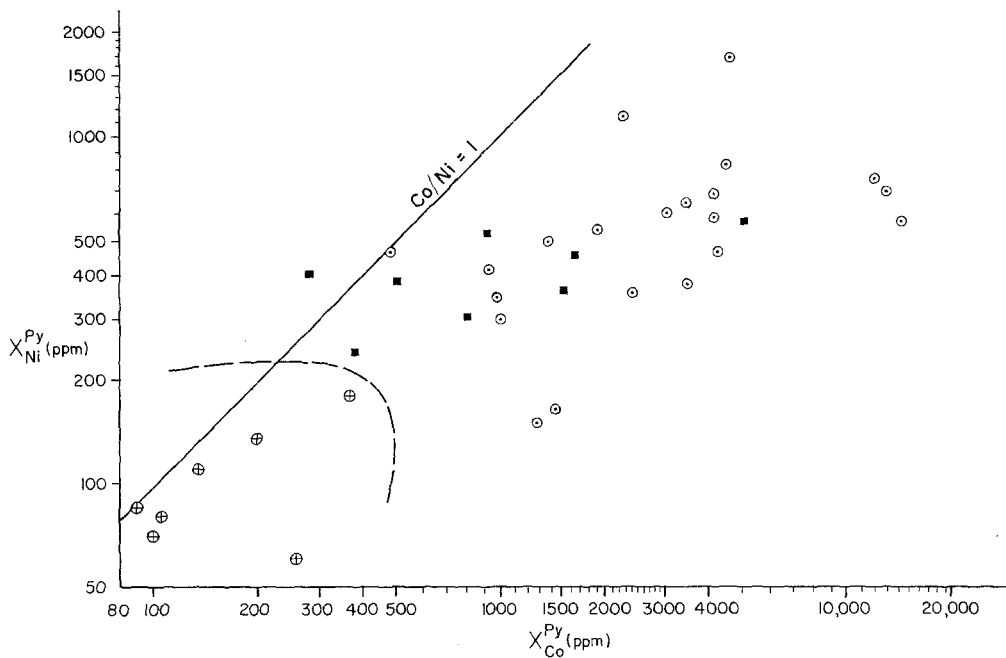
■ Disseminations and veinlets in metavolcanics

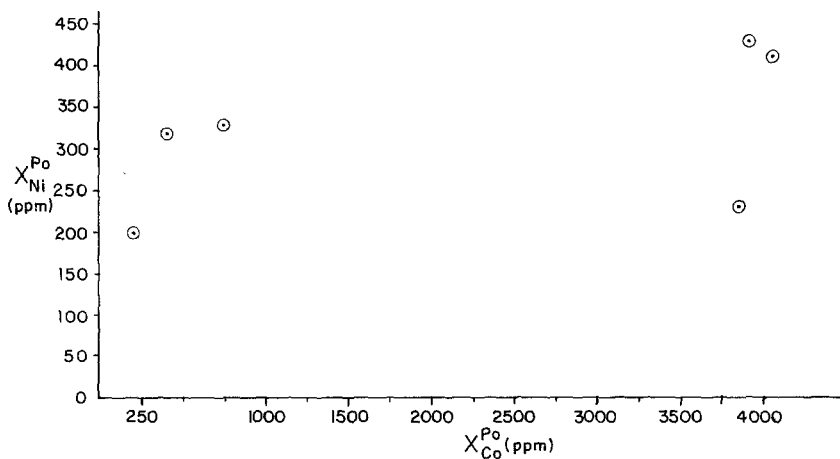Fig. 2. X_{Co}^{Py} Vs X_{Ni}^{Py} plot of analysed samples

Fig. 3. X_{Co}^{Po} Vs X_{Ni}^{Po} plot of analysed samples

Partitioning of elements between co-existing mineral pairs

Partitioning of nickel and cobalt in co-existing pyrite-pyrrhotite and pyrite-chalcopyrite pairs are shown in Fig. 5-8. Data on coexisting chalcopyrite-pyrrhotite pair are too meagre to be dependable. X_{Co}^{Py} Vs X_{Co}^{Po} (Fig. 5) shows a smooth linear variation. Sample 544 is ignored because of the high cobalt value where the constraint of a dilute solution is no longer

valid. The other three diagrams, i. e. the X_{Co}^{Py} Vs X_{Co}^{Cpy} plot, the X_{Ni}^{Py} Vs X_{Ni}^{Cpy} plot, and the X_{Ni}^{Py} Vs X_{Ni}^{Po} plot, are scattered.

The linear pattern can be tentatively accepted as having fulfilled the constraints of the dilute distribution law (e. g. Kern and Weisbrod, 1967) - i. e. the minerals are in equilibrium, Henry's Law is obeyed, and the solute substitutes for a solvent of the same charge. However, curvilinear or scattered distribution diagrams
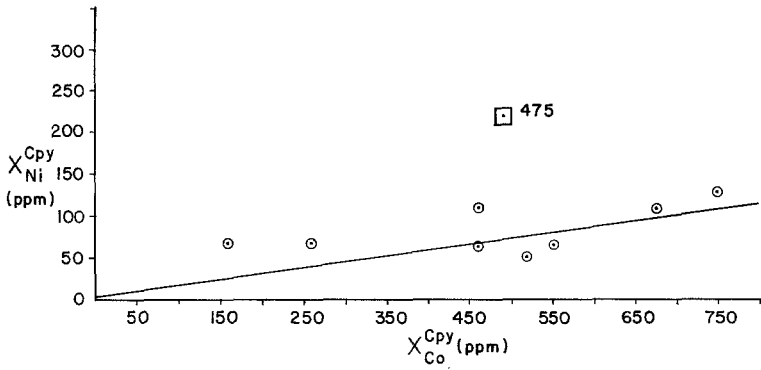
Fig. 4. X_{Co}^{Cpy} Vs X_{Ni}^{Cpy} plot of analysed samples
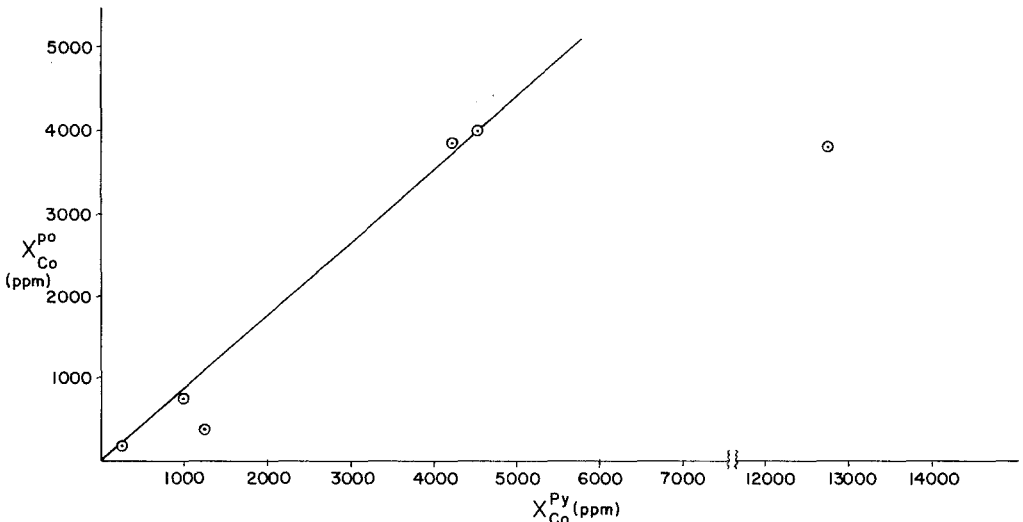
Fig. 5. X_{Co}^{Py} Vs X_{Co}^{Po} partition diagram

do not necessarily imply nonadherence to the constraints of the dilute distribution law; these may simply mean a varying P-T condition during mineralization such that the ratio of the activities of the element in the two phases changes systematically with P-T variation.

McIntyre (1958, 1963) has demonstrated that if substitution of an element in a struc ture necessitates formation of (induced) vacancies, the partitioning coefficient of the element - between the phase containing such induced point vacancies and another phase - would not follow the distribution law, even if the mineral-pair were in equilibrium and formed under near-constant P-T condition. Further, a second trace element, present in any one of the two phases, could also affect the distribution coefficient of the first (Kretz, 1959, 1960; Ghosh-Dastidar, 1969). In none of these two cases of nonadherence to the distribution law, a gross disequilibrium among the mineral pairs is indicated.

Ghosh-Dastidar et al. (1970) have derived relations to show that: (a) if the ratio X_i^β/X_i^α increases as X_i^α increases ("concentration plot"), the β phase contains induced vacancies accompanying the substitution of i : if the same ratio decreases as X_i^β increases, the induced vacancies are in the α phase. (b) If $R_i^{\alpha-\beta}$ is observed to be proportional to $(i/j)_\alpha$ ("interaction plot") i-j interaction in the α phase has affected the distribution of i between α and β.

In order to test whether the scattered distribution diagrams (Fig. 6, 7, 8) were due to disequilibrium condition or due to "induced vacancy" and/or "element interaction" phenomena, "concentration plots" and "interaction plots" were prepared.

Partitioning of cobalt between pyrite-chalcopyrite pair (Fig. 6) is found to be a scattered diagram. However, the partition-coefficient is observed to be dependent on cobalt concentration in pyrite (Fig. 9), thus indicating presence of induced point imperfections (which were the cause of departure from Henry's law) in chalcopyrite.

Partitioning of nickel between pyrite-pyrrhotite pair (Fig. 8) also produces a scattered diagram. However, when R_{Ni}^{Py-Po} is plotted against $(Co/Ni)_{Po}$ (Fig. 10) a straight line relationship is obtained. According to the model of Ghosh-Dastidar et al. (1970) this would indicate that cobalt-nickel interaction in pyrrhotite is responsible for the deviation from Henry's law.

When partitioning of nickel between chalcopyrite-pyrite pair is considered, induced vacancies in the chalcopyrite (Fig. 11) as well as Co-Ni interaction in chalcopyrite (Fig. 12) are both indicated as likely causes of deviation from Henry's law.

It is significant that within the pair chalcopyrite-pyrite, induced vacancies are indicated in the chalcopyrite phase accompanying substitutions of cobalt as well as nickel.

It is therefore argued that at least the mineral pairs pyrite-pyrrhotite and pyrite-chalcopyrite represent equilibrium assemblages, even though the partitioning coefficients of the trace metals in some cases give scattered diagrams. Departures from Henry's law, assuming deposition under near-constant P-T condition, are due to:
a) the presence of induced vacancies accompanying substitution of cobalt in chalcopyrite - in the case of partitioning of cobalt between pyrite - chalcopyrite;
b) the interaction of cobalt and nickel in pyrrhotite - in the case of partitioning of nickel between pyrite-pyrrhotite;
c) cobalt-nickel interaction, and presence of induced vacancies in chalcopyrite - in the case of partitioning of nickel between pyrite-chalcopyrite.

Further, systematic relationship between partition-coefficients and some crystal-chemical parameters (e. g. induced vacancies) implies that the trace elements occur in solid solution and not as adsorbed or occluded material in the sulphide phases. This, in its turn, would mean that higher trace-metal content of pyrite from the lodes is perhaps a reflection of the higher temperature of formation of the lodes.
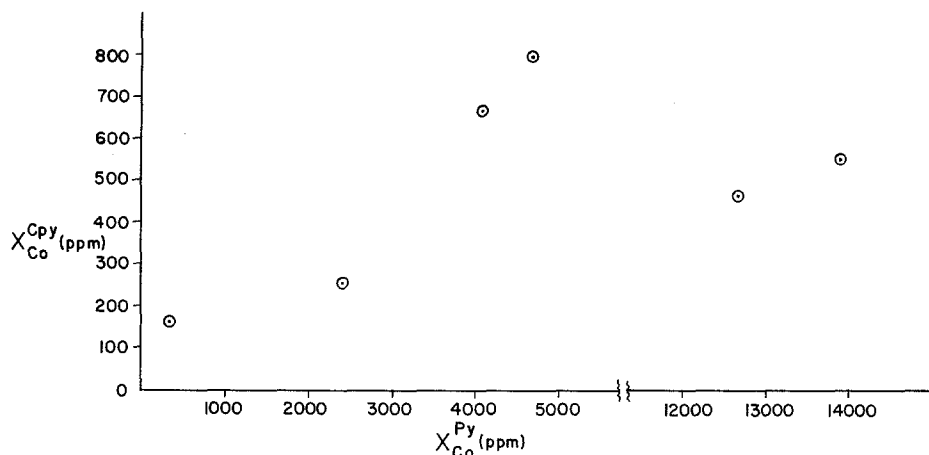
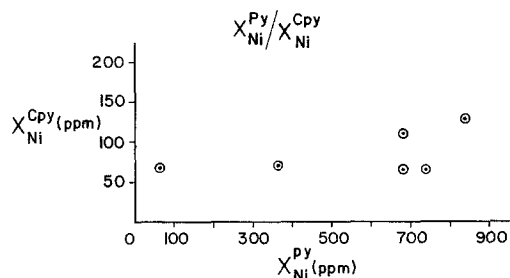Fig. 6. X_{Co}^{Py} Vs X_{Co}^{Cpy} partition diagram

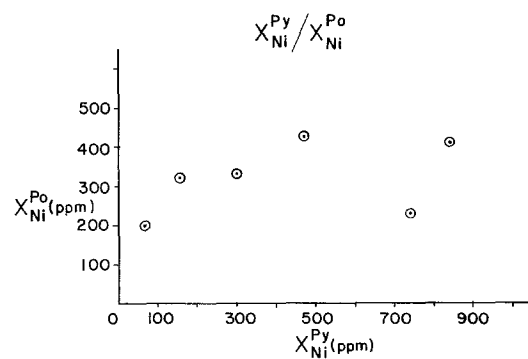Fig. 7. X_{Ni}^{Py} Vs X_{Ni}^{Cpy} partition diagram

Fig. 8. X_{Ni}^{Py} Vs X_{Ni}^{Po} partition diagram

Discussions

Co/Ni ratio in pyrite has been acclaimed by many workers as a sensitive indicator of (a) the environment of formation, (Davidson, 1962; Hegemann, 1943) (b) the source (Fleischer, 1955; Hawley, 1952, 1961), and/or (c) the degree of metamorphism suffered by the mineral deposits. In the last case, there is conflicting evidence, suggesting enhancement (Itoh, 1971a, b; Itoh and Kanehira, 1967) as well as depletion (Miyazaki et al., 1974) of the Co/Ni ratio with increasing metamorphism.

The Ingladhal suite of pyrite samples, as stated earlier, belongs to three different genetic groups: those belonging to the sulphide facies of iron formation; those belonging to the quartz-copper sulphide lodes; and those occurring as disseminations and veinlets in the metavolcanics. In the first group the tenors of Co and Ni are very low and the two trace metals occur in nearly equal proportion. In the second group the concentration levels of both trace elements are very high and the Co/Ni ratio ranges up to 27:1. The third group shows an intermediate range of cobalt concentration and very narrow range of nickel content.
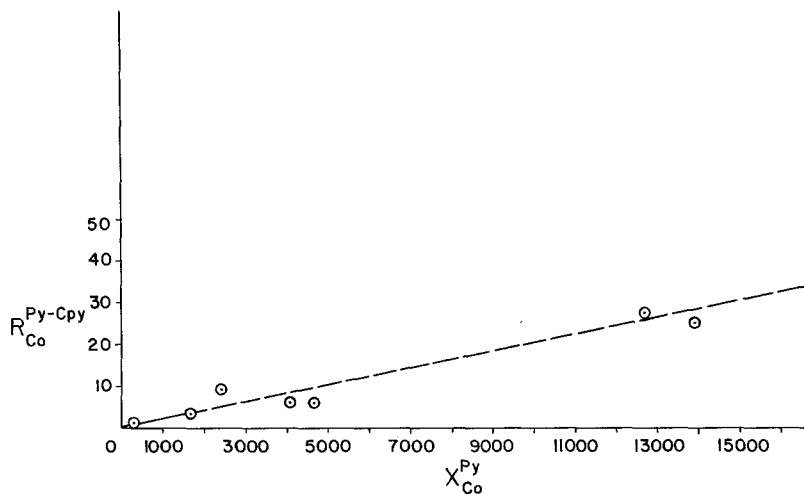
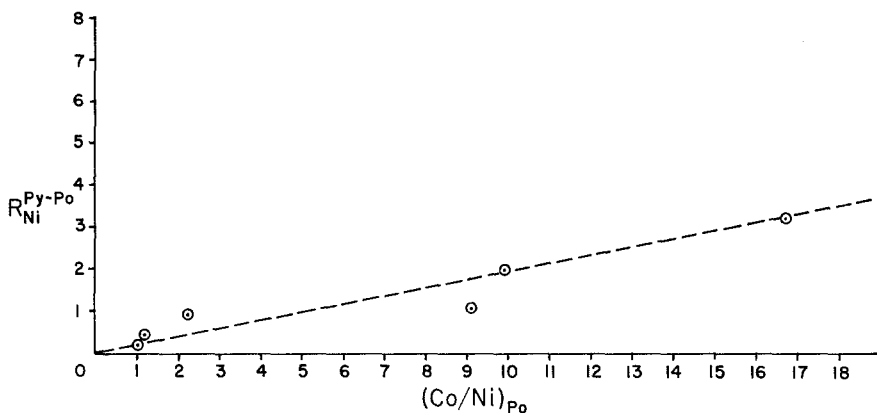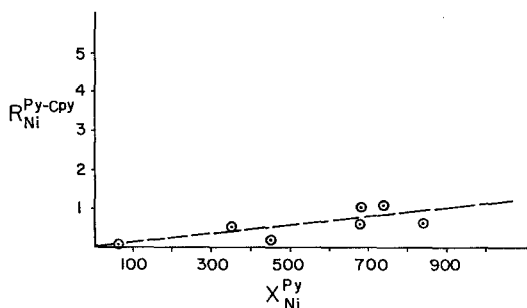Fig. 9. R_{Co}^{Py-Cpy} Vs X_{Co}^{Py} "concentration plot"

Fig. 10. R_{Ni}^{Py-Po} Vs $(Co/Ni)_{Po}$ "interaction plot"

Fig. 2 clearly brings out the difference be-
tween the first two groups. The low trace
metal content of the sedimentary pyrite
contrasts sharply with the high trace me-
tal content of pyrite from the quartz-
sulphide lodes. This clearly indicates
that within a given geological milieu
where the availability factor is presumab-
ly the same, trace metal content of py-
rite is mainly a function of the tempera-
ture of formation, especially when the
trace metals are found to be occurring in
solid solution. Pyrite disseminations in
metavolcanics, with variable cobalt con-

Fig. 11. R_{Ni}^{Py-Cpy} Vs X_{Ni}^{Py} "concentration plot"
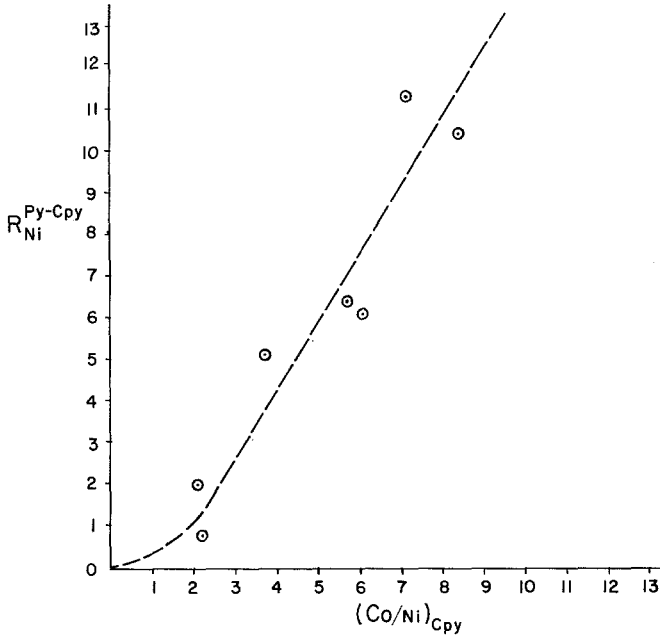
Fig. 12. R_{Ni}^{Py-Cpy} Vs $(Co/Ni)_{Cpy}$ "interaction plot"

tent and near-constant nickel values, per-haps signify metamorphic equilibration (of pyrite of diverse origin) whereby nickel is redistributed between coexisting sulphides and ferromagnesian minerals, while cobalt concentration in the pyrites remains practically unaffected (cf. Itoh, 1971). The only pyrite sample (448) with Co/Ni ratio less than one (0.67), shows sulphide dissemination confined within ejecta embedded in laminated tuff. This is taken to represent a rare instance of early magmatic immiscible sulphide liquid fraction, characterised by high nickel content (cf. Wilson, 1953). A chalcopyrite sample, (475) of similar mode of occurrence and presumably of the same origin, likewise shows high nickel value and lowest Co/Ni ratio among all analysed chalcopyrite samples.

Farkas (1973) concludes from his study of the Texas Gulf deposit that chalcopyrite tends to include Ni rather than Co in its structure. According to him, the fact that Co/Ni ratio is greater than unity for chalcopyrite in many instances, suggests that the initial ratio was very large in the ore fluid. In four out of six chalcopy-rite samples from Ingladhal, the Co/Ni ratios are lower than those in coexisting pyrite, implying a preferential enrichment of Ni in chalcopyrite. Cobalt and nickel contents in all chalcopyrite samples from the quartz-sulphide lode vary systematically (Fig. 4).

Where pyrite and pyrrhotite coexist, cobalt tends to be enriched over nickel in pyrite and Ni over Co in pyrrhotite (Vaughan, 1976), subject to the variation in local abundance of the elements. This is believed to be due to the greater crystal field stabilization energy of Co^{2+} which is low-spin in pyrite, as against high-spin Ni^{2+} which favours the pyrrhotite lattice. In three out of six analysed pyrite-pyrrhotite pairs, (nos. 74, 94 and 474) the nickel contents of pyrrhotite exceed those in coexisting pyrite.

In spite of the known preference for nickel (relative to cobalt) in chalcopyrite and pyrrhotite structures, not a single analysis of these two minerals from the lodes show Co/Ni < 1. This is taken to imply a very large Co/Ni ratio in the initial ore fluid. The discussion on the trace

element content of the host rocks (p. 3-5) also revealed that the volcanic rocks of this area are characterised by a higher cobalt content than their counterparts elsewhere. It is therefore tempting to suggest that the volcanic rocks as well as the different sulphide minerals at Ingladhal are all "stamped with the hallmark of a specific metal" (Williams, 1960), signifying consanguinity.

OREGENESIS IN THE LIGHT OF TRACE ELEMENT DATA

Modern theories of oregenesis in mineralized Archaean greenstone belts visualise ore-fluids as hot saline brines that may originate in diverse manners and at different stages of the geologic history of the terrane. The exhalative theory and its variants postulate ore deposition from volcanogenically-derived residual aqueous fluids at or near the volcanic rock/sea-water interface before or during diagenesis and lithification; the classical hydrothermal theory envisages a long time-interval between lithification of the host rock and ore deposition, from ore-fluids of same origin; yet another school (e. g. Anhaeusser et al. , 1969) visualises sulphide mineralization in Precambrian greenstone belts as a consequence of mobilization of chalcophile elements, initially present in the mafic lavas, through the agencies of metamorphism and metasomatism associated with younger granites.

Several aspects of trace metal distribution patterns in rocks and ores of the Ingladhal region assume special significance in this context.

First, a genetic connection between the volcanics and the ores is strongly indicated by the preponderance of cobalt in both.

Second, in barren metavolcanics, the occurrence of nickel and cobalt mainly in the silicate phase and that of copper mainly as sulphide seem to indicate an early separation of copper from cobalt and nickel.

Third, unusually high non-sulphide copper content of barren bedded cherts points to the availability of a copper-rich solution prior to their lithification.

Fourth, very high cobalt and nickel contents in orebody pyrite - in contrast with very low tenors of these trace metals in adjacent sedimentary pyrite - seem to imply a higher temperature of formation of the orebodies.

Mobilization of chalcophile elements in mafic lavas, through the agencies of metamorphism and metasomatism associated with younger granites, can perhaps be ruled out due to two reasons: radiometric age data [Ingladhal galena - 2900 ± 200 m. y. , Vinogradov and Tugarinov, 1968; Chitaldrug granite - 2400 - 2450 m.y. , Crawford, 1969] indicate that sulphide mineralization predates the nearby chitaldrug granite intrusion; and retention of very large amounts of nickel (and cobalt) in the silicate phase militiates against any hypothesis of efficient mobilization of chalcophile elements from the mafic lavas.

Mineralization at volcanic rock/sea-water interface appears unlikely, in view of the high temperature of formation of orebody pyrite - as reflected in its accommodation of large amounts of trace metals. Ore-fluid - seawater mixing models (Sato, 1972) visualise thermal equilibration between the two: temperature-salinity relationships determine whether precipitation would occur immediately and near the point of discharge, or somewhat later and away from the vent, - within depressions where physicochemical environment favours sulphide precipitation (e. g. where sulphide facies of iron-formation is being deposited).

Trace element data seem consistent with the model of a vocanoganically-derived Cu-rich ore fluid with inter-stratal migration below the sediment-water interface, and metallization prior to lithification of the volcaniclastic sediments (tuffs) and cherts. This would elegantly explain (a) retention of relatively high temperature of the ore-fluid (no mixing with seawater) and consequent

high trace metal accommodation in ore body sulphide minerals, (b) adsorption of appreciable amount of copper by the chert bands (availability of Cu-rich solution before lithification), and (c) stratiform nature of quartz-sulphide lodes within the tuffs.

SUMMARY AND CONCLUSIONS

1) The Ingladhal suite of metavolcanics is richer in cobalt and poorer in copper, compared to similar volcanic complexes elsewhere.

2) Among the metavolcanic complex, the tuffs are comparatively poorer in their nickel content, presumably due to subaqueous weathering of fine-grained ferromagnesian minerals of these volcaniclastic rocks.

3) About 90% of total nickel, 70% of total cobalt, but only 30% of total copper in the metavolcanics occur within the silicate phase. This implies a geochemical differentiation of copper from cobalt and nickel and its enrichment in a residual fluid.

4) Extremely low tenors of cobalt and nickel but high concentration of copper in the chert signify availability of such copper-rich solution before or during diagenesis of the cherts.

5) The quartz-sulphide lodes represent precipitation from such an ore-fluid in an environment conducive to sulphide formation (as is evident from the adjacent sulphide facies of iron formation).

6) Accommodation of very large amounts of trace metals within the lode sulphides - in contrast with the low tenors of trace metals in the sedimentary sulphides occurring in the same geologic milieu - is taken to indicate a higher temperature of formation of the lodes.

7) High temperature of formation and stratiform nature of the lodes, and high copper content of barren cherts imply interstratal migration of, and deposition from, the ore fluid. In other words,

mineralization is inferred to have taken place below the sediment-seawater interface without intermixing of ore fluid with seawater.

8) High cobalt content of metavolcanics and of the sulphide minerals in ore lodes are taken to signify a consanguinous relationship between the two.

9) Pyrites of sedimentary iron-formation, of volcanic-hydrothermal quartz-sulphide lodes, and of metamorphosed volcanics are characterised by distinctive patterns of Co-Ni distribution.

10) Partitioning of trace-metals between coexisting sulphide pairs in the ore bodies indicates that, in some instances, departures from Henry's law are due to induced vacancies and/or the interaction of a second trace metal in one of the mineral phases. Pyrite-pyrrhotite and pyrite-chalcopyrite minerals pairs represent equilibrium assemblages.

11) Very high copper content (≈ 500 ppm) of unmineralised chert bands, close proximity of orebodies with the sulphide facies of iron-formation, higher-than-usual cobalt content of the associated volcanic complex, and lower-than-usual copper content of these rocks seem to characterize the mineralized Ingladhal section of the larger Chitaldrug greenstone belt.

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