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Occurrence, chemistry and genesis of the nickel-rich phases in the ultramafic rocks from Swat, northwestern Pakistan

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ABSTRACT: *Trace amounts of finely disseminated Ni-rich phases, including heazlewoodite, pentlandite, millerite, godlevskite, violarite and awaruite, occur in the variably serpentinized ultramafic rocks of the Swat valley. The mode of occurrence, textural relationships with each other and with the associated silicate and oxide phases, relative modal abundance and chemical variation; all indicate that only the pentlandite represents the original sulphide phase. The rest are derived from pentlandite by the process of alteration. The additional amount of Ni, required for the conversion of pentlandite to heazlewoodite and other Ni-richer phases, was possibly derived by the serpentinization of primary olivine.*

INTRODUCTION

Disseminated grains of Ni sulphides (and arsenides) are fairly common in the altered (serpentinized/carbonated) ultramafic rocks of the alpine type (ophiolite) complexes (Ramdohr, 1967; Eckstrand, 1975; Lorand, 1985). Some of the earlier workers (e.g. Eckstrand, 1975) have postulated that serpentinization should upgrade the nickel content of sulphides in weakly mineralized ultramafic bodies so that a greater proportion of the total nickel content may be available for metallurgical recovery. Furthermore, progressive alteration of ultramafic rocks containing fine grained disseminations of Ni-rich phases may increase the volume of the mineralization and, therefore, result in the formation of massive ore deposits (Groves & Keays, 1979).

Accessory to trace amounts of Ni-rich phases also occur as fine grained disseminations in the variably altered ultramafic rocks of the Swat valley ophiolite (see also Chaudhry & Ashraf, 1986; Ashraf et al., 1989). A detailed investigation of the mode of occurrence, textural relationship and chemical composition of these phases, and a discussion about their genesis are the subject of the present study.

GENERAL GEOLOGY

The wedge-shaped zone of the Main Mantle Thrust (MMT), the locus of collision between the Indo-Pakistan and Kohistan island arc in the Swat valley, contains bodies of ophiolitic rocks, e.g. ultramafics, gabbros, pillow lavas and metasediments. These rocks occur as small to large lensoidal bodies distributed within and along the northern edge of the Indo-Pakistan plate. The ultramafic rocks, which dominate all the other lithologies of the different ophiolitic occurrences in the area, mostly consist of serpentinized harzburgites with only minor amounts of such types as dunite, lherzolite, websterite and pyroxenite. All the ultramafic rocks are invariably altered to varying degrees.

Although hydration leading to serpentinization is the principal type of alteration, locally however, carbonation has also taken place and drastically modified the original mineralogy of the ultramafic rocks investigated. The process of carbonation, which has produced different types of magnesite-rich rocks, post-dates that of serpentinization because veins of magnesite cutting across serpentine occur in several of the samples studied. At places (e.g. near the Barkotkai village), the serpentinized ultramafic rocks contain small bodies of chromitites and accessory to trace amounts of Ni-rich phases. Locally, the carbonate-rich assemblages in the study area contain emerald deposits which produce one of the world's finest gemstone quality emeralds (Fig. 1).

SAMPLES AND METHODS

A total of about 100 samples of the variably altered ultramafic rocks, including both the serpentinized and carbonated types, were collected from almost all the occurrences of such rocks in the area (Fig. 1). All the samples were studied petrographically and the ones containing grains of nickeliferous phases were selected for microprobe analyses and detailed SEM studies.

The microprobe analyses were performed at the Department of Geology, University of Leicester (UK), using Jeol Superprobe model JXA-8600. Analytical conditions were: 15 Kv accessory voltage; 30 x 10^{-9} probe current; 20 (2 x 10) seconds peak, 10 (2 x 5) seconds negative background and 10 (2 x 5) seconds positive background counting times. A focused beam (i.e. < 1 µm in diameter) of X-rays was used for analyzing the Ni-rich phases.

MODE OF OCCURRENCE AND PETROGRAPHY

Trace amounts of sulphide phase(s) occur as sparsely disseminated grains mostly in the main serpentine matrix of some of the rocks. However, some of the sulphide grains also occur in association with or within vein (original mesh line) magnetite. Although too small in most cases to be distinguished and recognized optically, they stand out prominently due to their high reflectance relative to the associated silicate and oxide phases. The studied sulphide phases, as characterized and distinguished on the basis of chemical composition, include heazlewoodite, millerite, pentlandite, godlevskite and violarite. Furthermore, grains of awaruite (an alloy of nickel and iron) were observed in some of the partly serpentinized samples. In most cases, the grains of awaruite are too small (less than 2 mm across) to be properly analyzed even with a focused beam of X-rays.

The various Ni-rich phases, mentioned above, constitute the following types of assemblages, all of which are accompanied by magnetite, Cr-magnetite and/or chrome spinel-ferritchromite associations: awaruite (e.g. sample B32), awaruite-millerite (e.g. sample L19), awaruite-heazlewoodite-millerite-pentlandite (e.g. sample L30), pentlandite-heazlewoodite-millerite (e.g. sample B26), pentlandite-heazlewoodite-millerite-godlevskite (e.g. sample B18), heazlewoodite-millerite (e.g. B29; heazlewoodite-millerite-godlevskite (e.g. sample L7), and pentlandite-violarite (e.g. sample Sp12).

Some of the rocks contain lamellar associations and intergrowths of pentlandite and heazlewoodite. In some cases, heazlewoodite seems to be replacing pentlandite. Conversely, pentlandite occurs as relict patches in some of the heazlewoodite-pentlandite composite grains. Locally, the pentlandite shows transformation to godlevskite. The close association of millerite with heazlewoodite probably suggests its development from the latter due to an increase in sulphur fugacity. The mode of occurrence,

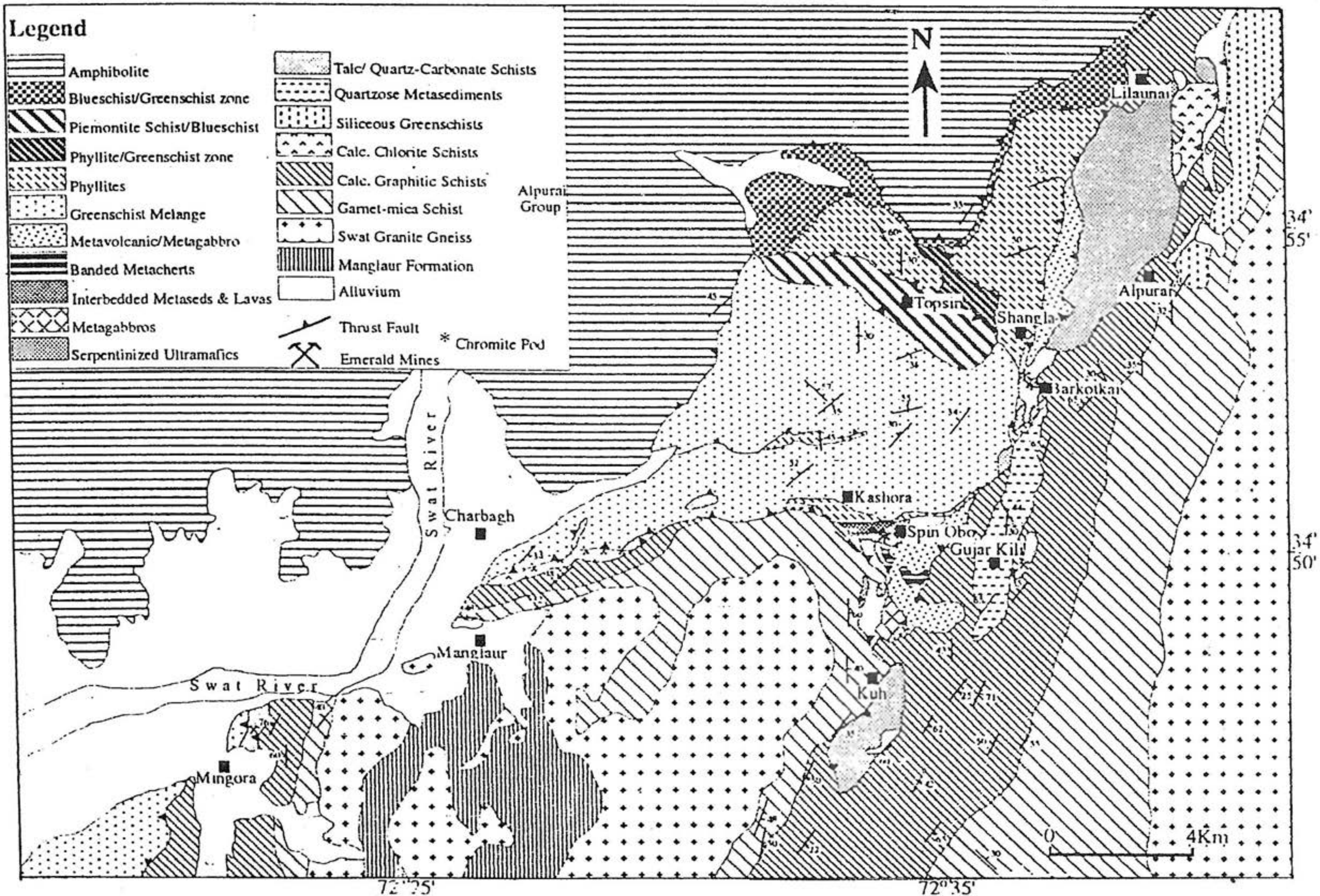


Fig. 1. Geological map of the Mingora-Lilaunai area, Swat (modified after Kazmi et al., 1984).

mineralogical associations, textural characteristics and alteration of the different Ni-rich phases have been illustrated in Figures 2, 3 and 4. The compositional characteristics of each of these phases are outlined in the following section.

PHASE CHEMISTRY

Representative analyses of all the investigated Ni-rich phases are listed in Table 1. Most of the sulphides analyses appear to be stoichiometric in terms of their M (= sum of the metals, i.e. Ni + Fe + Co + Cu): S (= sulphur) ratios (Fig. 5). Another peculiarity is that there is a perfect continuum (without any break whatsoever) between the most S-poor (i.e. phases with highest M:S ratios, e.g. heazlewoodite) and the most S-rich compositions (i.e. phases with lowest M:S ratios, e.g. millerite).

Pentlandite

Pentlandite was analyzed in ten of the studied samples. The metal (Ni + Fe + Co + Cu) to sulphur (S) ratio varies between 1.031 and 1.204 compared to 1.125 of the ideal pentlandite composition (M_9S_8). It means that some of the analyses are not stoichiometric due to either lower or higher sulphur contents than the stoichiometric value of 47.06 at. %. The amount of S in the analyses varies between 45.37 and 49.22 at. %. Although some appear to be slightly higher in S than the pentlandite reported by Misra and Fleet (1973), the S content of the studied pentlandites (except two analyses) closely matches the range observed by Harris and Nickel (1972).

Although the amounts of S and M:S ratios of the studied pentlandites depart from the ideal compositions, an almost 1:1 relationship between the S and M contents of the analyses suggest stoichiometry (see Fig. 6a). The ranges of Ni and Fe contents in the studied pentlandite are 23.94-34.95 at. % and 13.56-26.02 at. %, respectively. The amount of Co is highly variable (0.53-12.72 at. %) whereas that of Cu is mostly low (<0.07-1.55 at. %) (see Table 1). For being a part of a Ni-rich assemblage, the studied pentlandites lie on the Ni-richer side of the solid solution (Fig. 6b).

Heazlewoodite

Of all the sulphide phases occurring in the studied serpentinized ultramafic rocks, heazlewoodite is apparently the most abundant and well-formed. It was analyzed in 20 of the samples studied. The M:S ratios of the analyses (1.403-1.604) are very similar to that demanded by the stoichiometric composition (M_3S_2; M:S = 1.5). The amount of S varies from 38.00 to 41.59 at. % whereas the ranges in the contents of the different metals are: 52.62-60.98 at. % Ni; <0.05-4.67 at. % Fe; <0.05-6.26 at. % Co; and <0.07-0.83 at. % Cu (Table 1). The overall range and amount of Co appear to be large for heazlewoodite, however, Co in the studied heazlewoodite is mostly low ($\leq \sim 1$ at. %) except for two analyses. The range of Fe in the analyses is also higher than what has been previously reported for heazlewoodite (see Misra & Fleet, 1973). It should, however, be noted that both the Co and Fe vary systematically with, and apparently replace, Ni in the structure (Fig. 7). These findings might indicate that larger amounts of Co and Fe, if available, can be accommodated in the structure of heazlewoodite.

Millerite

The composition of millerite was determined in fourteen samples. The M:S ratio of the analyses varies between 0.945 and 1.191. Some of the analyses are S-deficient relative to the stoichiometric composition (NiS). The two major components, i.e. Ni and S, range from 46.34 to 53.65 at. % and 45.64 to 51.39 at. %,

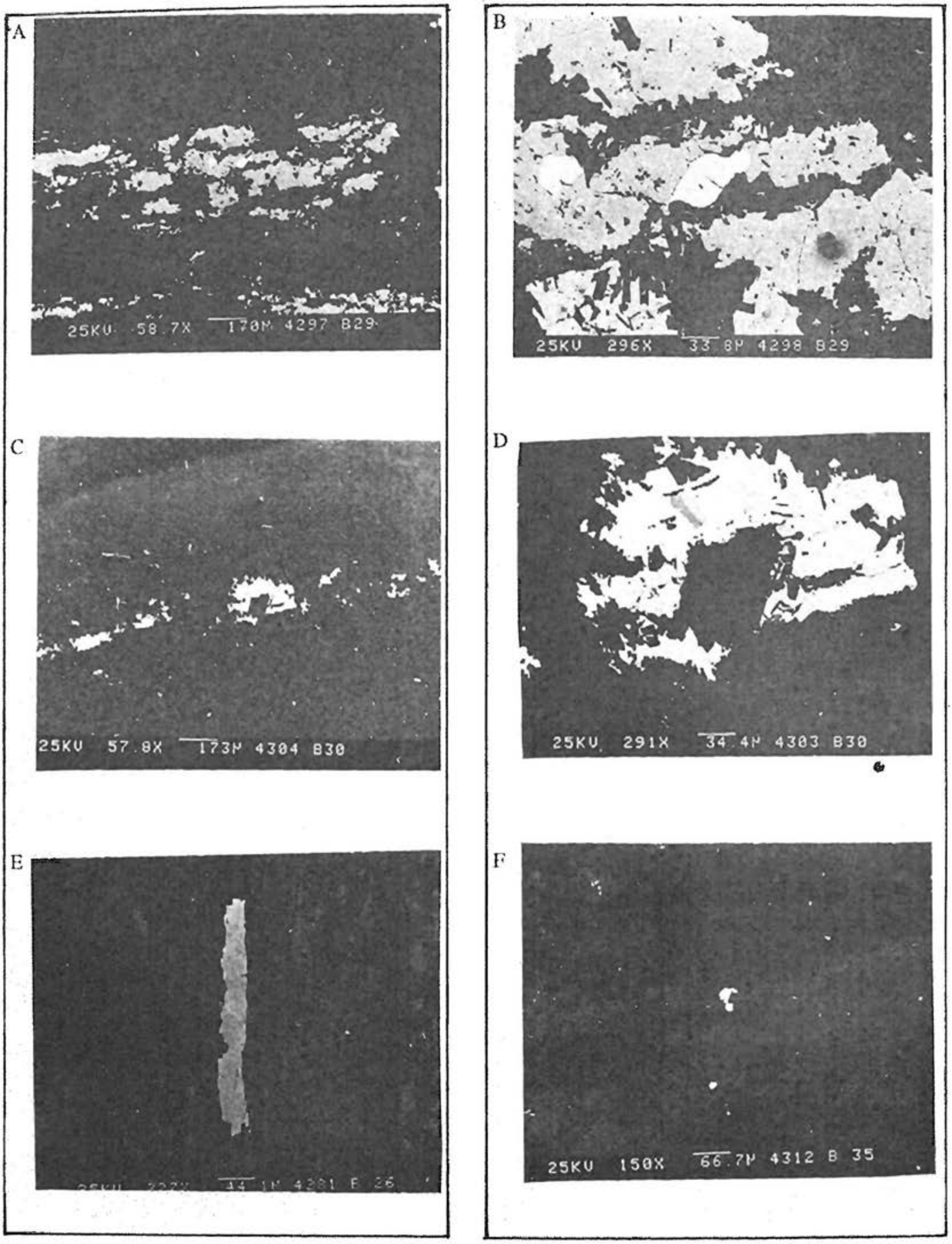


Fig. 2. SEM photographs: (A) grains of heazlewoodite (bright) along vein (original mesh line) magnetite (grey); (B) magnified view of the central part of (A); (C) heazlewoodite along mesh line magnetite; (D) magnified view of the central part of (C) showing that heazlewoodite (bright) also encloses magnetite; (E) elongated pentlandite (grey)-heazlewoodite (light grey) composite grain along a fracture in a coarse grained olivine (black); (F) a small grain of awaruite (bright) associated with recrystallized olivine (grey) that occurs along a fracture in one of the samples studied.

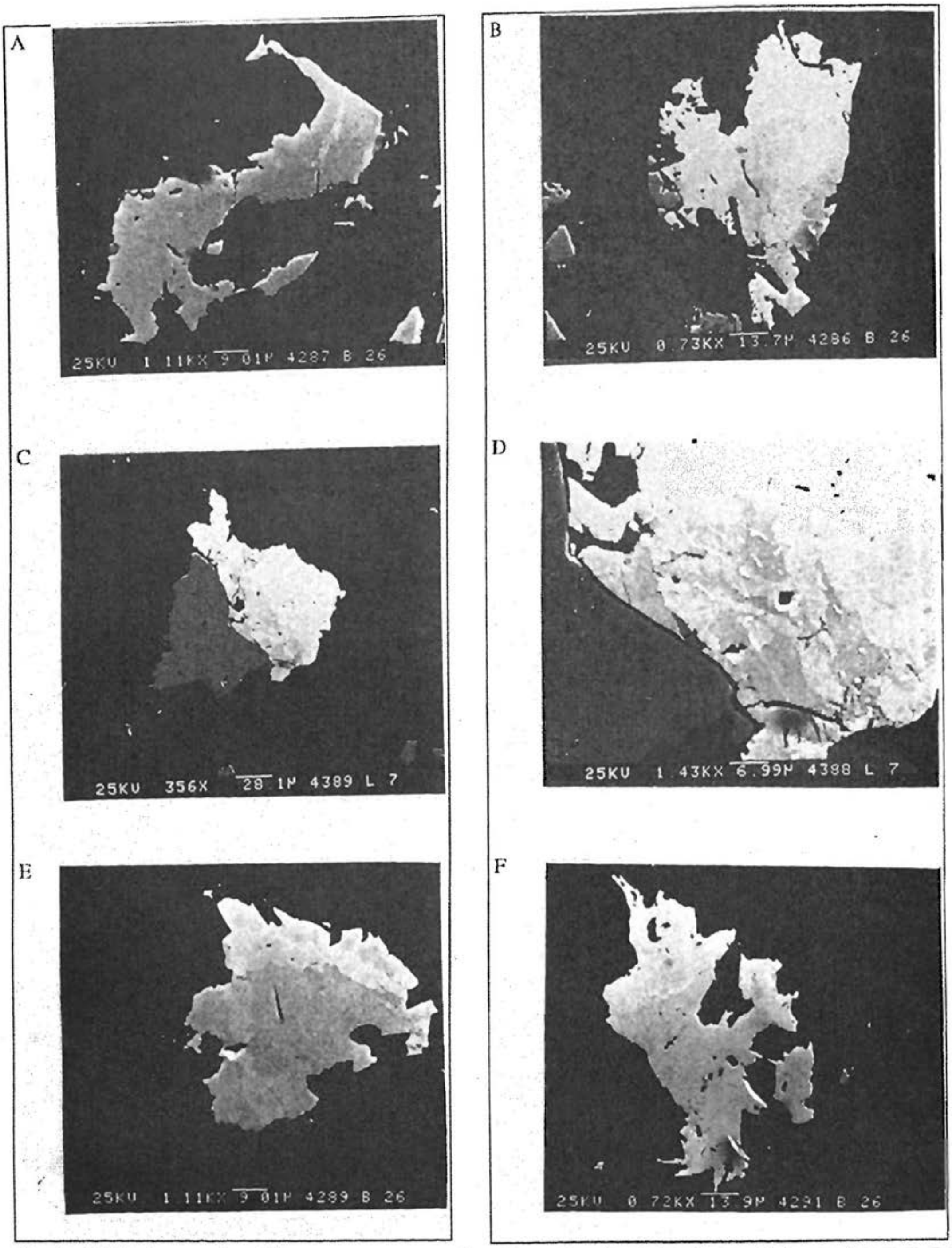


Fig. 3. SEM photographs: (A) a composite grain of heazlewoodite (bright)-millerite (light grey) associated with ferritchromite (grey); (B) heazlewoodite (light grey)-pentlandite (grey) lamellar association; (C) magnetite (probably exsolved) in a bastite grain (black background) intergrown with godlevskite (bright) as shown in the magnified view (D); (E) heazlewoodite (lighter grey)-pentlandite (grey) lamellar association; relic patches of pentlandite are visible in the lamella of heazlewoodite as can be seen in the magnified view (F).

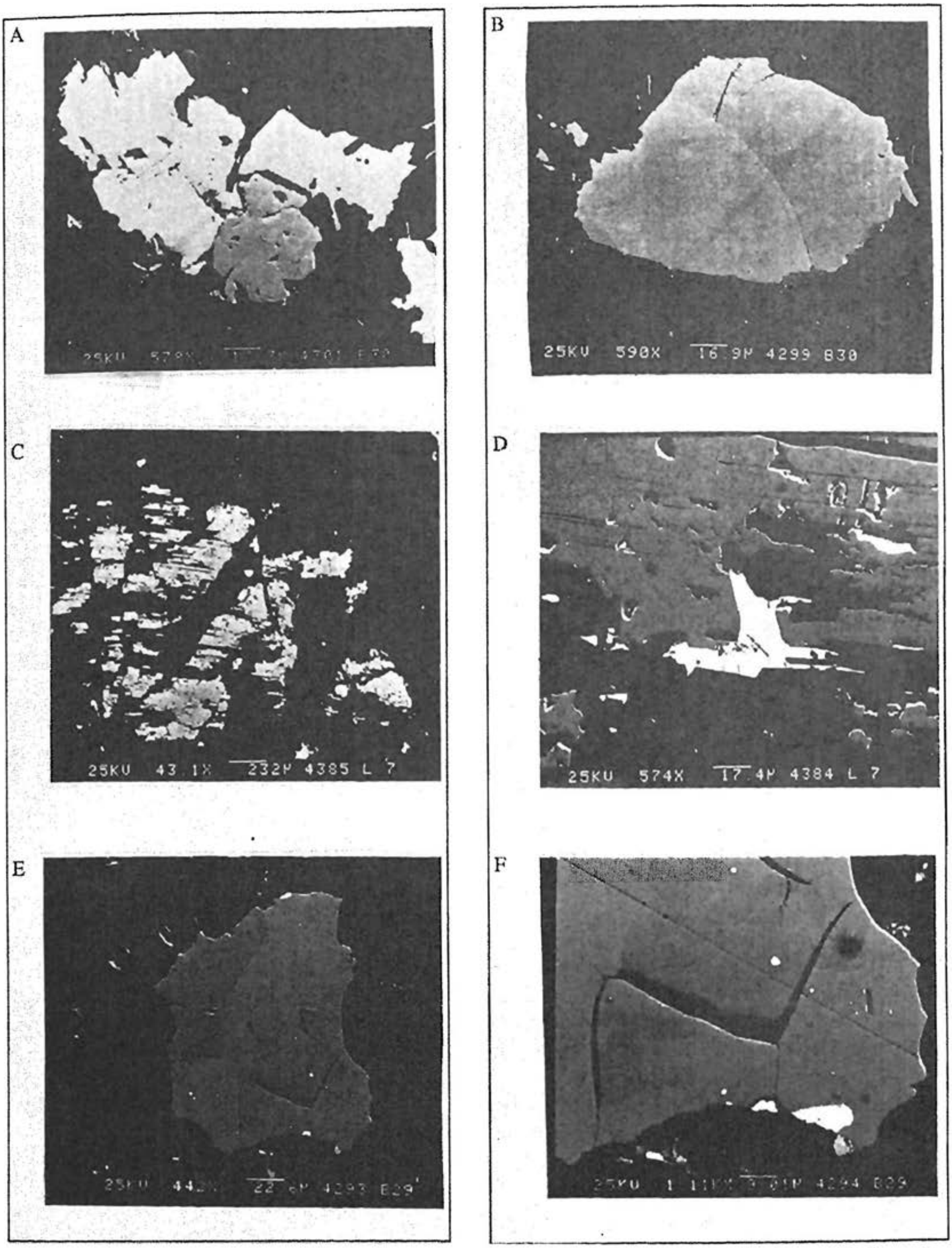
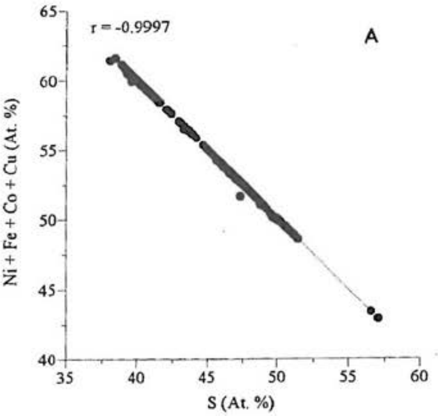


Fig. 4. SEM photographs: (A) and (B) heazlewoodite (light grey) replacing pentlandite (grey); (C) lamellar association of magnetite (dark grey) and heazlewoodite (bright grey), the latter contains relic patches of pentlandite (grey) as shown in the magnified view (D); (E) a composite grain of heazlewoodite (light grey) and pentlandite (grey), the former contains relic patches of pentlandite and conversely, heazlewoodite replaces pentlandite in the latter; (F) a composite grain consisting of pentlandite (darker grey relic patches) altered to heazlewoodite (bright grey) and godlevskite (grey).



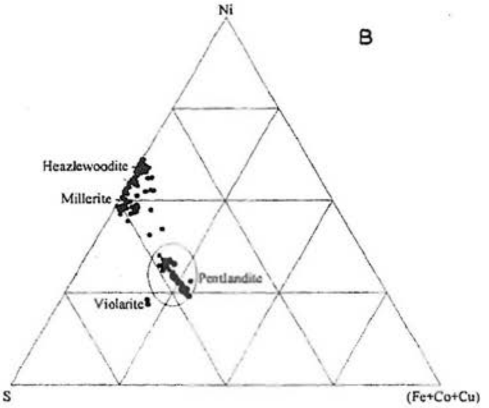
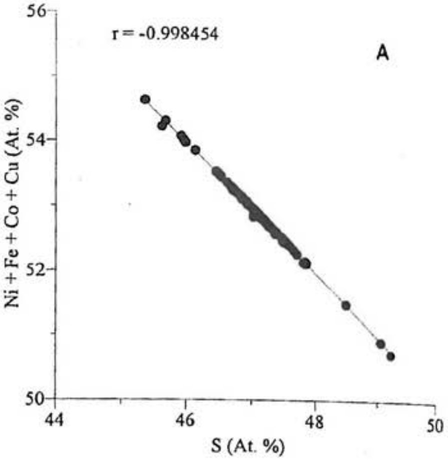


Fig. 5a. Relationship between the metal and sulphur contents of all the sulphides studied.

5b. Classification of the sulphide species in terms of S, Ni and Fe+Co+Cu.

respectively. The total amount of 'foreign' metals (other than Ni) is low in most of the analyses (≤ ~ 2 at. %): 0.08-2.01 at. % Fe; <0.05-1.79 at. % Co; and <0.07-1.57 at. % Cu (Table 1). Furthermore, all the analyses contain small amounts of As which in some cases range up to 1 at. %. This suggests that a minor amount of S has been replaced by As and ignoring this replacement in the calculation of M:S ratios may, at least partly, be responsible for the seemingly non-stoichiometric character of some of the analyses. Inter-element relationships in the analyses show that all of the 'foreign' metals substitute for Ni in the structure of millerite (see Fig. 8).



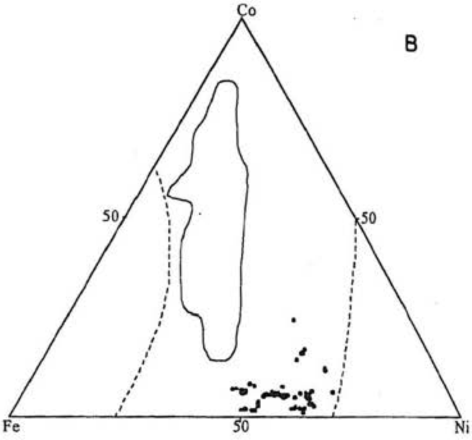


Fig. 6a. Relationship between the metal and sulphur contents in pentlandite.

6b. Compositional characteristics of pentlandite. Th dashed lines mark the limits of maximum amounts of Fe and Ni in natural pentlandites (Knop & Ibrahim, 1961). The solid line outlines the compositional field of pentlandite from the upper part of the upper zone in the western Bushveld complex (taken from Merkle & von Gruenewaldt, 1986).

Godlevskite

Godlevskite (ideally M_7S_6 with a M:S ~ 1.167; where M = Ni + Fe + Co + Cu), analyzed in two samples, has a M:S ratio ranging from 1.132 to 1.164. The ranges in the contents of the various components in the studied godlevskite

TABLE 1. REPRESENTATIVE ANALYSES (ATOMIC PERCENT) OF THE NICKELIFEROUS OPAQUE PHASES.

Samp.	B32		L19			L30					
Phase[1]	Aw	Aw	Aw	Aw	Mil	Aw	Aw	Pn	Pn	Hzw	Hzw
Ni	84.07	83.57	82.44	74.45	53.02	74.13	81.84	24.19	24.43	59.68	58.86
Fe	13.80	14.71	13.54	24.46	0.08	24.44	14.23	25.27	26.31	0.26	1.03
Co	0.29	0.36	0.41	0.46	0.02	0.65	0.45	3.69	2.18	0.02	0.02
Cu	1.83	1.33	3.58	0.61	0.11	0.74	3.47	0.01	0.05	0.04	0.08
S	0.00	0.02	0.02	0.00	46.76	0.02	0.00	46.82	47.02	39.99	40.01
As	0.01	0.01	0.01	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.00
Sb	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Ratio[2]	5.28*	5.10*	4.70*	2.92*	1.138	2.87*	4.51*	1.136	1.126	1.50	1.50
Samp.	L30				B26						B29
Phase[1]	Hzw	Hzw	Mil	Mil	Pn	Pn	Hzw	Hzw	Mil	Mil	Hzw
Ni	59.46	59.33	48.28	48.16	29.27	33.00	58.99	58.84	48.45	48.51	58.23
Fe	0.41	0.50	0.93	2.00	14.55	13.56	0.80	0.70	0.28	0.31	1.91
Co	0.01	0.02	0.00	0.10	8.93	6.37	0.07	0.07	1.22	1.16	0.02
Cu	0.08	0.05	0.06	0.08	0.01	0.01	0.07	0.04	0.01	0.07	0.07
S	40.03	40.09	50.71	49.53	47.22	47.04	40.06	40.34	50.01	49.92	39.70
As	0.01	0.01	0.01	0.11	0.00	0.01	0.01	0.01	0.03	0.03	0.07
Sb	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Ratio[2]	1.50	1.49	0.972	1.016	1.118	1.126	1.50	1.48	0.999	1.003	1.51
Samp.	B29			B18							
Phase[1]	Hzw	Hzw	Mil	Pn	Pn	Hzw	Hzw	Mil	Mil	Gsk	Gsk
Ni	58.81	57.74	50.35	29.49	29.18	59.99	59.08	48.42	49.73	48.843	47.684
Fe	1.06	2.52	1.40	15.22	15.18	0.09	0.45	0.27	0.09	3.749	4.165
Co	0.05	0.02	0.05	8.30	8.54	0.03	0.06	0.03	0.17	1.040	1.229
Cu	0.07	0.05	0.05	0.01	0.01	0.09	0.06	0.16	1.57	0.088	0.073
S	39.98	39.66	48.13	46.93	47.05	39.76	40.33	51.09	48.38	46.241	46.804
As	0.03	0.01	0.02	0.04	0.02	0.04	0.01	0.03	0.04	0.031	0.037
Sb	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.008	0.008
Ratio[2]	1.50	1.52	1.077	1.130	1.125	1.51	1.48	0.957	1.066	1.162	1.136
Samp.	L7							Sp1		Sp12	
Phase[1]	Hzw	Hzw	Mil	Mil	Gsk	Gsk	Gsk	Pn	Pn	Vl	Vl
Ni	59.62	59.54	47.81	48.68	48.24	48.86	48.59	33.81	33.01	23.00	21.71
Fe	0.10	0.12	1.82	0.91	4.22	3.85	3.64	17.39	18.68	18.28	20.71
Co	0.04	0.02	1.30	1.38	1.24	0.94	1.08	1.67	0.99	2.09	0.46
Cu	0.24	0.03	0.08	0.04	0.03	0.10	0.06	0.13	0.06	0.01	0.00
S	39.98	40.23	48.81	48.77	46.18	46.20	46.56	46.98	47.25	56.60	57.10
As	0.02	0.05	0.18	0.21	0.08	0.04	0.06	0.02	0.01	0.02	0.01
Sb	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Ratio[2]	1.50	1.48	1.045	1.046	1.163	1.164	1.146	1.128	1.116	0.766	0.751

[1] Aw = awaruite, Gsk = godlevskite, Hzw = heazlewoodite, Mil = millerite, Pn = pentlandite, Vl = violarite

[2] Denotes sum of metals (Ni + Fe + Co + Cu) divided by sulphur, except for awaruite (the analyses marked with asterisks), where the ratio is equal to Ni divided by ΣM (= Fe + Co + Cu)

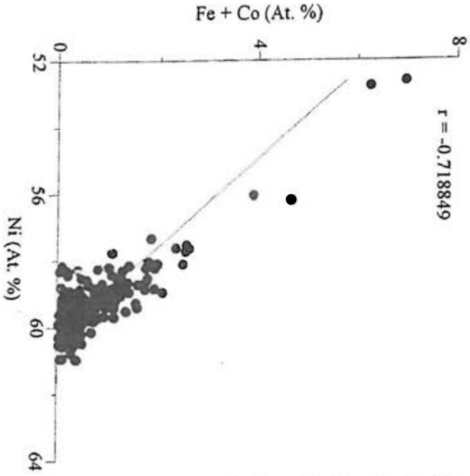


Fig. 7. Relationship of Ni with Co+Fe in heazlewoodite.

are: 47.68-48.86 at. % Ni; 3.64-4.22 at. % Fe; 0.94-1.24 at. % Co; <0.07-0.10 at. % Cu; and 46.18-46.88 at. % S (see Table 1).

Violarite

This phase was observed as relic patches within grains of garnierite in a completely serpentinized sample (Sp12). Based on averaging the two analyses presented in Table 1, the studied violarite contains 22.36 at. % Ni, 19.50 at. % Fe, 1.28 at. % Co, and 56.85 at. % S. The resulting M:S ratio (0.759) closely approximates the value of 0.750 based on the stoichiometric composition $[(FeNi)_3S_4]$. Its mode of occurrence, i.e. association with pentlandite within garnierite (a weathering product), suggests that it probably formed due to the alteration of pentlandite (see also Misra & Fleet, 1974).

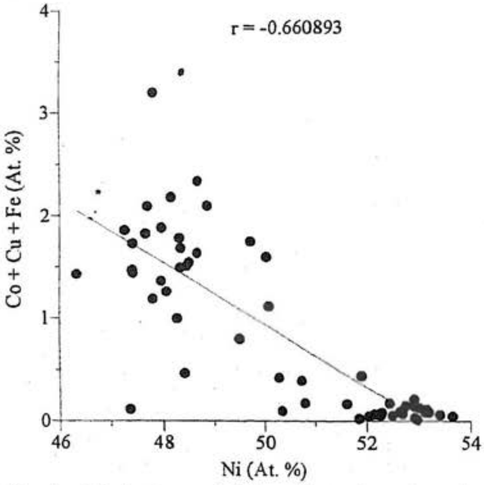


Fig. 8. Variation of Ni with other metals in millerite.

Awaruite

Awaruite, an alloy mainly consisting of Ni and Fe, was analyzed in seven samples. The ranges of its different components are: 71.89-87.10 at. % Ni; 8.6-26.0 at. % Fe; <0.06-2.01 at. % Co; and 0.13-4.97 at. % Cu. The Ni to Fe ratio varies between 2.76 and 10.12 whereas the ratio between Ni and the sum of the other three metals (Fe, Co and Cu) ranges from 2.56 to 6.78 (Table 1). The previously reported general range for the Ni:Fe ratios in awaruite is 1:1 to 4:1 (see Ashley, 1975) and is believed to increase with the degree of serpentinization (Lorand, 1985). An earlier conception regarding the Co concentration in awaruite (Ahmed & Bevan, 1981; Ahmed & Hall, 1982) seems to be also true for those of the present study. That is, the presence of appreciable Co (> 1 at. %) is restricted to analyses with less than 78 at. % Ni. The Ni content of awaruite is believed to be related to the type of assemblage: awaruite coexisting with heazlewoodite only is distinctly richer in Ni (~ 70-80 at. %) than that coexisting with troilite-pentlandite (~ 62 at. % Ni) (Misra & Fleet, 1973). The present studies evidently support this earlier observation, however, some of the studied awaruites are distinctly enriched in Ni (containing up to 87 at. % Ni) compared to the previously reported compositions (Ahmed & Hall, 1982). The compositional characteristics and relationships in awaruites from the studied rocks are illustrated in Figure 9.

Olivines in six out of the seven awaruite containing samples was found to be distinctly

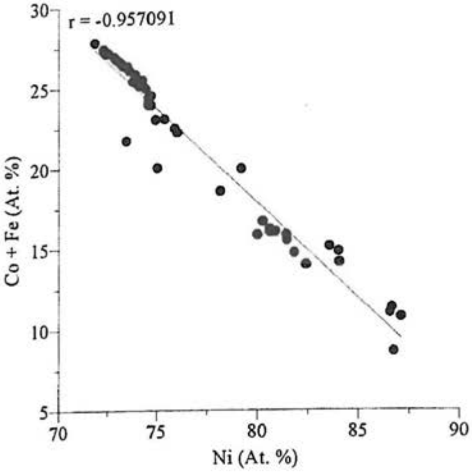


Fig. 9. Relationship of Co+Fe with Ni in awaruite.

less magnesian, i.e. Fo content ≤ 90, than that in samples lacking awaruite (see Arif, 1994). This suggests that the distribution of awaruite is related to the Mg content of olivine or, conversely, the Fo content of olivine somehow depends on the absence or presence of awaruite in the host rock.

DISCUSSION

Before, during and, perhaps also, after emplacement, the ultramafic rocks underwent more or less pervasive serpentinization. This resulted in almost complete conversion of original olivine to serpentine and the formation of bastites at the expense of almost all the original orthopyroxene grains. The serpentinization-related expansion resulted in the formation of inter- and intra-granular cracks and fractures in the rocks.

In general, ultramafic rocks commonly contain trace to accessory amounts of sulphide phases occurring as blebs that probably originated as immiscible droplets. This is also the case with most of the ultramafic rocks from the study area. However, it seems that the present state and mineralogical composition of the nickeliferous opaque assemblage in the studied rocks are largely a result of extensive modification of their original sulphide component. One strong indication in this regard is that heazlewoodite is much more abundant and well-formed than pentlandite. The reverse would be the case had the sulphide assemblage been not modified. This means that, relative to unmodified purely magmatic sulphide assemblage, the sulphide component in the studied rocks is much more Ni-rich. The formation of Ni-rich liquids in nature does not seem possible and almost all the demonstrably magmatic sulphide occurrences have compositions that lie within the MSS (mono-sulphide solution) at Fe/Ni > 1, most having Fe/Ni between 1.5 and 10 (cf. Groves et al., 1974). In contrast, sulphides in the studied rocks, whether considered individually as separate phases or collectively as assemblages, are characterized by negligibly low Fe/Ni ratios. Furthermore, textural relationships suggest that only pentlandite represents the original sulphide phase whereas all the others, especially heazlewoodite and millerite, are derived from pentlandite through serpentinization. In other words, the Ni content of the original magmatic sulphide component of the studied ultramafic rocks has been upgraded. The additional amount of Ni was probably provided by its release from the primary silicate(s), i.e. olivine, during serpentinization. The alternative explanation that Ni was added from an external source is not plausible because there are no indications that the host rocks are enriched in this metal (see Arif, 1994).

In addition to variable amounts of Fe, small quantities of Ni, Co and even of Cu can also enter into the structure of olivine (cf. Thalhammer et al., 1986). Therefore, not only Fe but significant amounts of Ni and Co can be expected to be released during the serpentinization of olivine. This is because serpentine, unlike its precursor (olivine), cannot accommodate more than only a small fraction of these ore-forming elements. Furthermore, the amount of Ni, so set free, may increase substantially with progressive serpentinization and metamorphism (Donaldson, 1981). The magnetite, Cr-magnetite and ferritchromite – all occurring as zones/rims around, or enclosing, relics of Cr-rich spinel – in the studied ultramafic rocks are strongly enriched in Ni (containing up to 2.03 wt % NiO) (see Arif, 1994). All these phases are undoubtedly secondary in origin (i.e. non-magmatic) and their formation is most probably related to serpentinization and/or metamorphism of the host rocks. It clearly demonstrates that alteration has liberated Ni, most probably from olivine, the only silicate phase that undoubtedly has undergone serpentinization. The relatively low Ni content of the serpentines studied (see Arif, 1994), further supports the release of this metal during serpentinization. As Ni is strongly chalcophilic, its high concentration in magnetite, Cr-magnetite and ferritchromite may represent only a small fraction of that actually released. In other words, originally a much larger amount of Ni was set free, most of which was used up in upgrading the already existing sulphide component, and only the remainder was adjusted into magnetite and/or ferritchromite.

CONCLUDING STATEMENT

The pervasive serpentinization of the rocks resulted in the almost complete obliteration of primary olivine and transformation of the original orthopyroxene. Besides producing inter- and intra-granular cracks, the process of serpentinization resulted in the liberation of abundant Ni, Fe and Mn which, especially the last two, were consumed in the formation of Cr-magnetite and/ or ferritchromite. The major bulk of the released Ni was, however, utilized in upgrading the Ni content of the original sulphides and producing new Ni-rich phases. In other words, the present assemblage of nickeliferous phases in the studied rocks is largely the result of serpentinization.

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