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Garnierites from the ultramafic rocks in Swat, northwestern Pakistan

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ABSTRACT: *The serpentinized and/or carbonated ultramafic rocks of the Swat valley ophiolite locally contain accessory to trace amounts of microscopic to megascopically visible grains of garnierite. Such grains mostly occur in samples where the serpentinisation and/or carbonation are rather pervasive and accompanied by supergene alteration. Their optical properties and chemical compositions are extremely variable and differ greatly from spot to spot within individual grains which appear to be intimate mixtures of different compositions. The highly variable and, in most cases, high to very high concentration of FeO (up to ~54 wt.%), and abnormally high CaO contents (ranging up to ~3 wt.%) in the chemical analyses make them distinct from the serpentine varieties of garnierites reported from other localities.*

Most of the garnierite grains seem to be pseudomorphic after, and contain relic patches of, pentlandite, and might have developed as a result of reaction between the 'normal' matrix serpentine or talc and the original magmatic sulfide. As their occurrence along fractures in the rocks is clear at least in some cases, their formation probably took place after the main episodes of serpentinisation and carbonation and was probably brought about by the processes of weathering and supergene alteration. The association of calcite with garnierites in some of the rocks indicates that the fluid, which caused the alteration, was charged with calcium and CO_2 and this factor probably also accounted for the anomalously high values of CaO in the garnierites.

INTRODUCTION

The alteration of ultrabasic rocks results in the formation of nickeliferous laterite and nickeliferous serpentine deposits. The nickel is concentrated into a large number of phyllosilicates whose intimate physical as well as interlayered mixing is clearly exhibited on X-ray diffraction patterns (Hotz, 1964) and has been confirmed by high resolution electron microscopy (Uyeda et al., 1973). It is therefore difficult to obtain monomineralic phases by mechanical separation (Manceau & Calas, 1985) and that is why a generalized term 'garnierite' is used to describe collectively these mixtures of nickel-containing hydrous silicates.

Brindley and Hang (1973) carried out a broad survey of garnierites and concluded that they are usually serpentine group minerals or talc-like minerals or mixtures of these. The studies of Springer (1974), however, suggest that nickel-bearing varieties of sepiolite, chlorite, vermiculite and even the amorphous and more hydrated forms of these hydrous nickel-containing silicates also occur. This is in agreement with the Faust's (1966) conclusion, based on a review of garnierites, that five types of minerals comprise the garnierite group, namely nickel-bearing varieties of serpentine, chlorite, talc, sepiolite and pimelite.

Nickel-containing minerals in the serpentine group resemble lizardite and chrysotile (Brindley & Wan, 1975). A nickel analogue of chrysotile has been named pecoraite (Faust et al., 1969) and the name nepouite is given to a nickel analogue of lizardite (Maksimovic, 1973; Brindley & Wan, 1975). Although a nickel analogue of the six-layer orthoserpentine (Brindley & Von Knorring, 1954; Zussman & Brindley, 1957) has been synthesized by Jasmund and Sylla (1971a, b), a nickel analogue of antigorite, used in the strict sense of a mineral with a long super lattice 'a' parameter, has not been observed. The nickel analogue of kerolite (10 Å talc) is designated as pimelite (Maksimovic, 1973) which is considered to be a hydrated form of willemseite - the nickel analogue of talc (Springer, 1974). A nickel-bearing chlorite has been named nimite (De Waal, 1970).

The present study is based on petrographic and mineralogical observations performed on specimens of garnierites which occur in the pervasively altered ultramafic rocks of the Swat valley ophiolite, northwestern Pakistan. The chemical analyses of these specimens mostly conform to that of serpentine intimately mixed on a submicroscopic scale. However, the amounts of Fe and Ca in these analyses are distinctly high compared to those previously reported from other occurrences (e.g. Springer, 1974; Brindley & Wan, 1975; also see Newman & Brown, 1987, for a review). An examination and understanding of these anomalies might lead to a further extension in the existing compositional limits of garnierites. Furthermore, the studied samples are perhaps the only examples of garnierites which contain well-preserved relics of their precursors.

GENERAL GEOLOGY

The Main Mantle Thrust (MMT), which is the locus of collision between the Indo-Pakistan plate and Kohistan island arc in NW Pakistan, is marked by the occurrence of a zone of melanges in the Swat valley. Based on the nature of the principal or diagnostic component lithology, Kazmi et al. (1984) distinguished these melanges into three main types: (1) blueschist melange, (2) greenschist melange and (3) ophiolitic melange.

The ophiolitic melange consists of a series of lensoidal bodies of mafic-ultramafic rocks distributed along the northern edge of the Indo-Pakistan plate in Swat. The occurrence of such bodies is best exemplified by the outcrops in and around the Barkotkai and Spin Obo villages (Fig. 1). The various lithological units in these areas, comprising different types of ultramafic rocks containing small pods of chromitite, gabbros, basaltic-andesitic pillow and amygdaloidal lavas, as well as metamorphosed maganiferous quartzose sediments and cherts, are representative of a typical ophiolitic complex.

The ultramafic rocks of the ophiolitic melange are invariably altered (serpentinized and carbonated) to varying degrees. The sequential alteration of the ultramafic rocks has, at places, given rise to a zonal pattern within individual bodies consisting of a weakly serpentinized core, surrounded by an extensively developed outer zone of completely serpentinized rocks and a locally present peripheral shell of talc-magnesite ± quartz lithologies. Similarly, the mafic plutonic and volcanic rocks are altered and metamorphosed under the upper greenschist to lower amphibolite facies conditions such that their original mineralogy is completely obliterated.

SAMPLES AND METHODS

A total of about 50 samples of the variably altered ultramafic rocks, including both the serpentinized

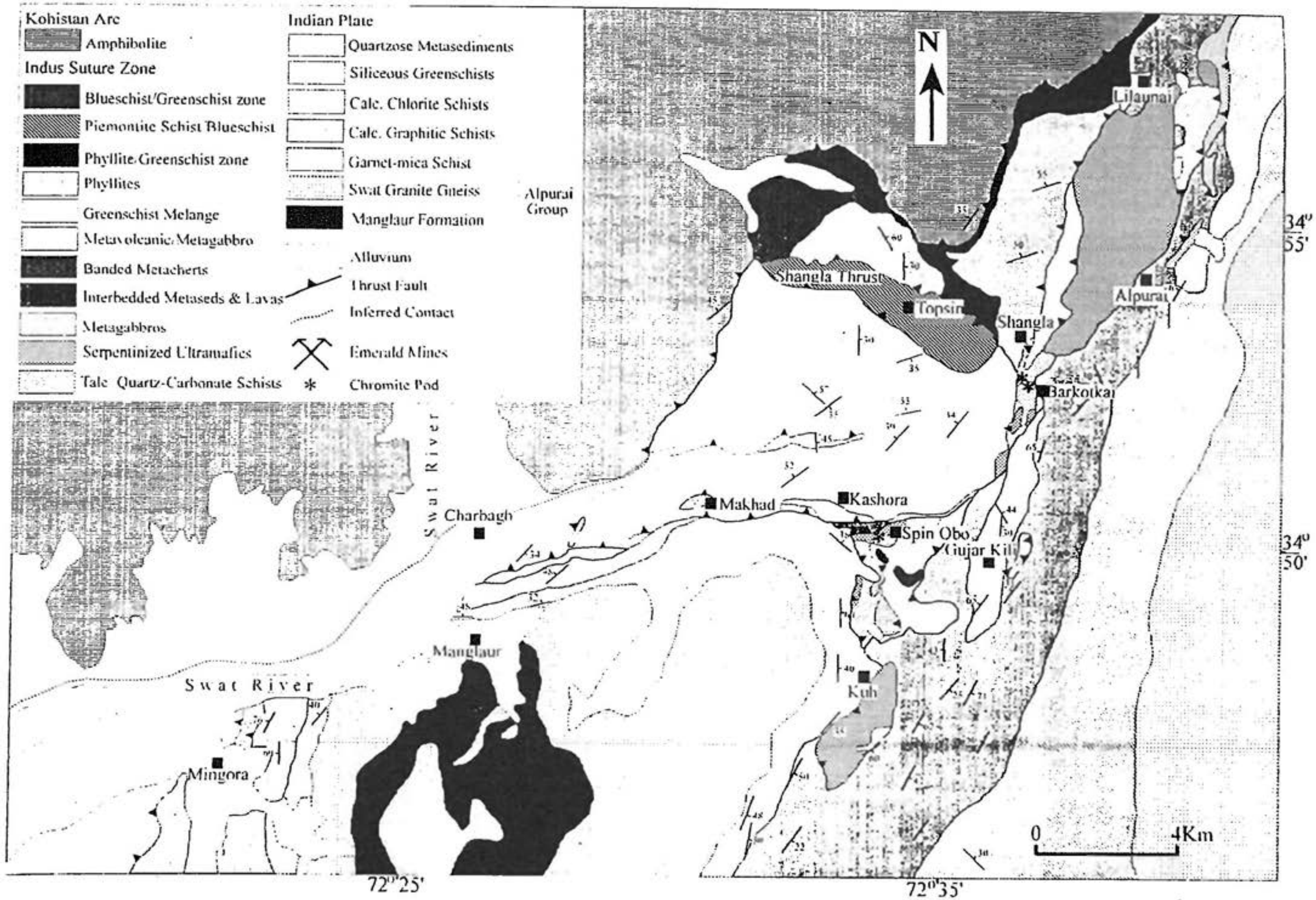

Fig. 1. Geological map of the Mingora-Lilaunai area of Swat, northwestern Pakistan (modified after Kazmi et al., 1984).

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 garnierites 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 accelerating 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. An X-ray beam of less than 1 µm diameter was used during the analysis of the garnierite grains due to their intragranular compositional heterogeneity. The patches of pentlandite occurring as relics within the garnierite grains were also analysed with a focused (< 1 µm diameter) beam.

PETROGRAPHY

The grains of garnierites occur mostly in the completely serpentinized ultramafic rocks of the area (Fig. 1). These rocks are so altered that, except for some rare relic cores of chrome spinel grains, none of the primary magmatic phases can be observed in them. That is why serpentine and magnetite are the only principal constituents of most of these rocks. As the pseudomorphic (mesh and curtain) textures are totally lacking in these rocks, the process of serpentinisation probably took place under relatively higher temperature conditions (cf. Wicks & O'Hanley, 1988). Although most of the magnetite in the rocks could be the product of serpentinisation of the original mafic silicate phases, i.e. olivine and pyroxenes, some of the magnetite grains contain patches of chrome spinel suggesting their development partly at the expense of the magmatic spinel grains. In some cases, the small, relic, grey, solid cores of chrome spinel are separated from the outer bright magnetite by a highly reflectant, porous zone of ferritchromite. Rarely, very minor amounts of muscovite and calcite also occur in these rocks. The mode of occurrence and cross-cutting relations of muscovite and calcite with serpentine indicate that their formation probably commenced after the main episode of serpentinization. The occurrence of these phases also suggests that potash and lime were probably introduced into the rocks after their serpentinization. In addition, grains of garnierites also occur, although less commonly than in the serpentinites, in the carbonated varieties of the ultramafic rocks which contain abundant magnesite and talc or quartz with or without traces of Cr- spinel.

The garnierites occur in accessory amounts (<0.5 modal %) as small, microscopic to megascopically visible grains of brown colour distributed mainly along fractures in the rocks. In transmitted light, they exhibit a yellowish brown to dark brown colour with a reddish birefringence. Their colour also varies within individual grains, some of which even contain patches that are opaque in transmitted light. In reflected light, the grains of garnierite stand out prominently in the rock due to their much high reflectance relative to the silicate and/or carbonate matrix (Fig. 2). However, the individual grains exhibit an intragranular variation in reflectance which is consistent with the difference in transmitted light properties. This heterogeneity in chemical composition, indicated by the within-grain variation in colour, birefringence, and reflectance, was confirmed by microprobe analysis.

Opaque patches in some of the nickel-containing silicate grains range from 3 to more than 15 µm in diameter and are identified as pentlandite on the basis of microprobe analysis. This suggests that the main precursor to the

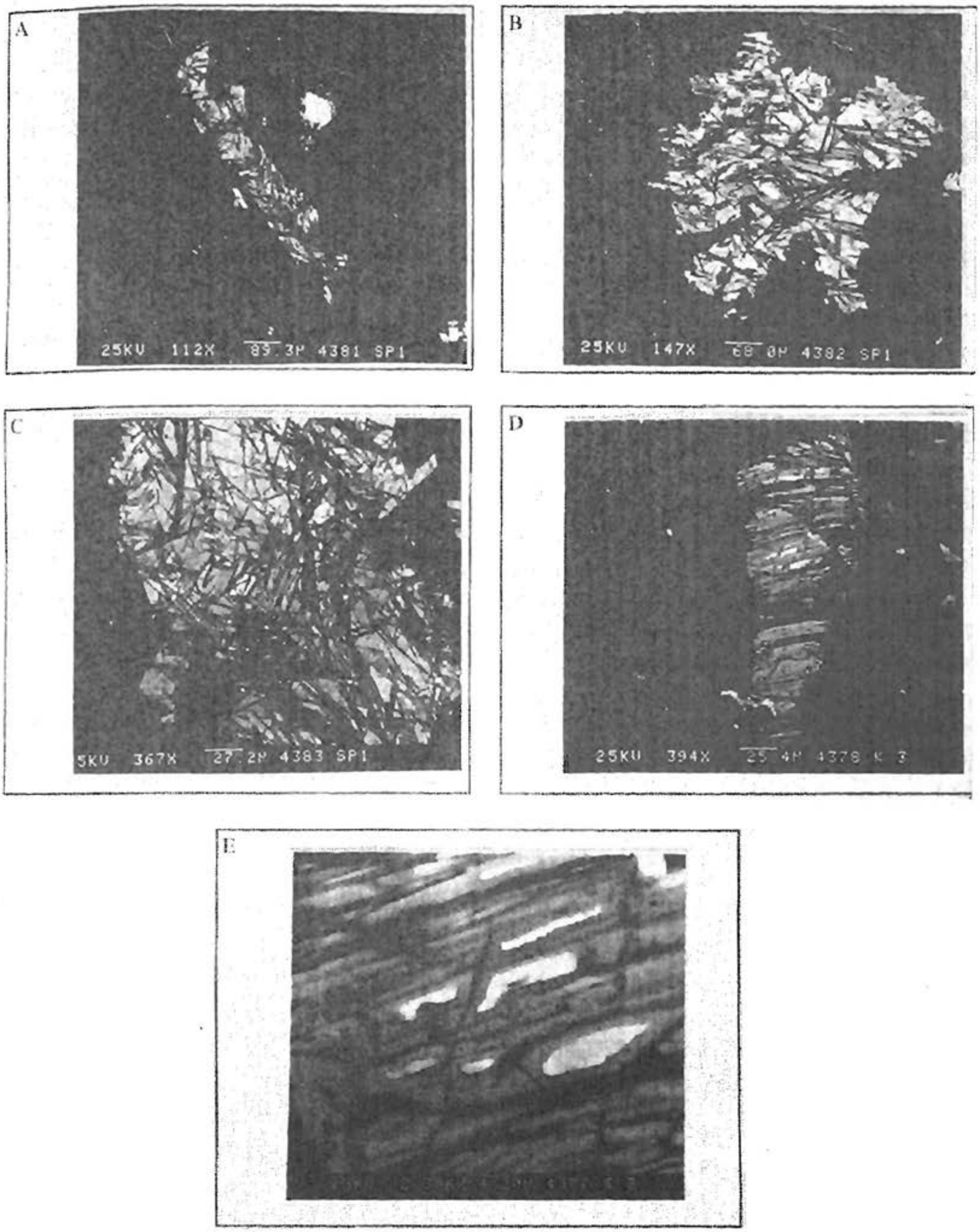

Fig. 2. Back-scattered electron images showing grains of garnierite containing relic patches of sulfide(s) (bright white) which are readily visible in D and in its magnified view E.

Ni-containing phases was probably pentlandite that acted as a source for the Ni and Fe contents in garnierites. The overall form, general habit and cleavage patterns, detectable in some of the grains, also suggest that garnierites are pseudomorphic after the primary magmatic pentlandite. It should, however, be noted that some of the opaque patches within the grains of garnierites are of magnetite probably representing a product of oxidation of some of the iron of the original sulfide grains.

PHASE CHEMISTRY

Garnierites

Grains of these Ni-containing silicate(s) were analysed in seven serpentinized and two carbonated ultramafic rocks. The composition of the garnierites is extremely variable in terms of all the major components even within individual grains. The analyses consist principally of four oxides and the concentration of each of them varies between broad limits (Table 1). The overall variation in the chemistry (in wt. %) of the studied garnierites can be summarised as follows: SiO_2 = 16.08-46.41; TiO_2 = <0.06-0.07; Al_2O_3 = <0.02-3.90; Cr_2O_3 = <0.06-0.76; FeO* = 0.50-53.51; MnO = <0.06-0.14; MgO = 0.14-37.99; CaO = 0.05-2.8; Na_2O = <0.02-0.70; K_2O = <0.03-0.73; and NiO = 0.95-41.83. Recalculation on the basis of seven oxygens yield about two Si and a sum of approximately three octahedral cations for many of the analyses. It implies that, although being highly enriched in Ni, Fe and Ca, the majority of the analyses closely approximate the composition of serpentine. However, there are several analyses with wt.% SiO_2 and totals too high for serpentines. In addition, some of the analyses, especially the ones that are high both in total iron and nickel, have the sum of ions, calculated on the basis of seven oxygens, in excess of five and the number of Si atoms less than two. Some of these analyses might be closer to talc in composition with totals being low due either to (1) a higher water content than 'normal' talc, i.e. they represent a hydrated form of talc, or (2) the assumption that all the iron is present as FeO, i.e. some of the iron could actually be in the form of Fe_2O_3. The possibility that some of the compositions could be talc cannot be ruled out because of their association and/or intergrowth with the 'normal' flaky talc.

To verify whether all of the analyses represent serpentine or some are actually talc, is not important because of extreme heterogeneity in composition within individual grains, nonetheless, the studied garnierites have two diagnostic chemical features which have not been previously reported from elsewhere (see Table 1). These are:

1. Their FeO content is extremely variable and, in many cases, abnormally high (reaching up to ~ 54 wt. %).
2. Many of them contain rather high amounts of CaO (reaching up to ~ 3 wt. %) showing systematic trends with the other octahedral cations. In contrast, all but one (analysis 11, containing 1.07 wt. % CaO; Brindley & Wan, 1975) of the previously reported analyses on Ni-rich serpentines (Ni-lizardites, nepouites and brindleytes; see analyses quoted by Springer, 1974; Brindley & Wan, 1975; Maksimovic & Bish, 1978; Newman & Brown, 1987) contain very little or no Ca.

The validity of the analyses is indicated by an almost 1:1 relationship between Si and the sum of the octahedral cations (Fig. 3a). The conclusion (based on petrographic observations and chemical characteristics) that the studied garnierites may include both the serpentine and talc varieties is confirmed by plotting them on Springer's (1974) diagram (Fig. 3b).

TABLE 1. REPRESENTATIVE ANALYSES OF GARNIERITES

Samp.[1]	B1	K	K	K	L1	Sp	Sp	Sp	Sp	Sp	Sp	Sp	Sp
SiO_2	41.58	39.68	32.21	31.37	38.57	43.06	41.68	38.65	39.71	37.17	36.23	39.92	36.65
TiO_2	0.00	0.01	0.01	0.03	0.02	0.02	0.02	0.01	0.01	0.01	0.01	0.01	0.01
Al_2O_3	0.76	0.02	0.15	0.01	0.11	0.64	0.48	0.71	0.62	0.32	0.46	0.44	0.34
Cr_2O_3	0.76	0.05	0.11	0.01	0.11	0.21	0.23	0.43	0.27	0.13	0.25	0.12	0.15
FeO*	4.17	42.48	34.75	41.92	3.71	4.48	10.88	11.31	5.53	14.44	13.23	9.45	13.65
MnO	0.05	0.01	0.03	0.03	0.07	0.06	0.02	0.05	0.08	0.07	0.06	0.08	0.07
MgO	37.62	2.42	7.95	2.09	1.61	37.99	31.58	30.28	33.05	25.76	26.82	29.13	26.13
CaO	0.05	2.08	1.09	1.30	0.54	0.05	1.49	0.36	0.10	0.54	0.57	0.38	0.34
Na_2O	0.01	0.19	0.09	0.11	0.07	0.01	0.00	0.01	0.01	0.00	0.01	0.02	0.00
K_2O	0.05	0.57	0.18	0.42	0.06	0.00	0.03	0.00	0.01	0.02	0.02	0.03	0.01
NiO	1.73	2.67	6.43	7.30	41.16	1.79	2.09	3.80	5.20	7.02	9.00	10.41	10.08
Total	86.78	90.18	83.00	84.59	86.03	88.31	88.56	85.61	84.59	85.48	86.66	89.99	87.43
Cations per seven (O, OH)[2]													
Si	1.986	2.248	2.008	2.021	2.310	2.017	2.025	1.972	1.998	1973	1.913	1.982	1.929
Ti	0.000	0.000	0.000	0.001	0.002	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.001
Al	0.042	0.001	0.011	0.001	0.007	0.035	0.027	0.043	0.037	0.020	0.029	0.026	0.021
Cr	0.028	0.002	0.005	0.001	0.005	0.008	0.009	0.017	0.011	0.005	0.010	0.005	0.006
Fe^{2+}	0.166	2.013	1.811	2.259	0.186	0.175	0.442	0.483	0.233	0.641	0.584	0.392	0.601
Mn	0.002	0.000	0.002	0.002	0.004	0.002	0.003	0.002	0.003	0.003	0.003	0.003	0.003
Mg	2.679	0.204	0.739	0.201	0.144	2.652	2.288	2.303	2.479	2.039	2.111	2.156	2.050
Ca	0.002	0.126	0.073	0.090	0.035	0.003	0.078	0.020	0.005	0.031	0.032	0.020	0.019
Na	0.002	0.021	0.011	0.014	0.009	0.001	0.000	0.001	0.001	0.000	0.001	0.002	0.000
K	0.004	0.041	0.014	0.035	0.005	0.000	0.002	0.000	0.001	0.001	0.001	0.002	0.001
Ni	0.067	0.122	0.322	0.378	1.983	0.067	0.082	0.156	0.210	0.300	0.382	0.416	0.427
Total	4.977	4.778	4.996	5.003	4.688	4.961	4.956	4.997	4.978	5.013	5.066	5.004	5.057

(Continued)

Samp.[1]	Sp	Sp	Sp	Sp	Sp	Sp	Sp	Sp	Sp	Sp	Sp	Sp	Sp1
SiO_2	29.73	31.81	32.12	33.92	32.25	43.45	34.57	33.60	41.96	46.41	28.71	40.56	41.58
TiO_2	0.01	0.01	0.02	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.05	0.01
Al_2O_3	0.08	0.14	0.05	0.27	0.01	0.08	0.01	0.02	0.00	0.00	0.00	3.31	1.26
Cr_2O_3	0.09	0.08	0.07	0.17	0.01	0.03	0.07	0.05	0.04	0.01	0.04	0.39	0.26
FeO*	34.55	25.51	37.06	27.29	35.44	11.93	32.09	23.61	8.11	9.13	15.58	3.01	7.30
MnO	0.05	0.07	0.03	0.06	0.04	0.03	0.03	0.08	0.12	0.07	0.05	0.00	0.07
MgO	5.85	12.97	3.74	10.16	0.98	8.14	0.84	3.54	2.32	2.20	0.95	37.24	34.36
CaO	1.80	1.18	1.704	1.53	1.92	1.66	1.99	1.34	1.53	1.39	1.03	0.11	0.08
Na_2O	0.01	0.01	0.07	0.08	0.15	0.01	0.07	0.04	0.09	0.02	0.01	0.08	0.02
K_2O	0.02	0.02	0.05	0.05	0.08	0.01	0.03	0.03	0.04	0.03	0.02	0.13	0.01
NiO	11.29	13.59	14.65	16.63	20.92	21.84	22.26	25.03	26.78	32.15	34.48	1.15	1.23
Total	83.48	85.39	89.56	90.18	91.81	87.19	91.97	87.35	80.73	91.42	80.88	86.03	86.18
						Cations per seven (O, OH)[2]							
Si	1.920	1.904	1960	1.951	1.967	2.365	2.060	2.063	2.493	2.476	1.993	1.935	2.018
Ti	0.001	0.000	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.002	0.000
Al	0.006	0.010	0.004	0.018	0.001	0.005	0.001	0.001	0.000	0.000	0.000	0.186	0.072
Cr	0.005	0.004	0.003	0.008	0.000	0.001	0.003	0.002	0.002	0.000	0.002	0.015	0.010
Fe^{2+}	1.866	1.277	1.891	1.313	1.808	0.543	1.599	1.212	0.406	0.407	0.905	0.120	0.296
Mn	0.003	0.004	0.002	0.003	0.002	0.001	0.002	0.004	0.006	0.003	0.003	0.000	0.003
Mg	0.563	1.158	0.340	0.871	0.089	0.661	0.075	0.324	0.207	0.175	0.098	2.648	2.486
Ca	0.125	0.076	0.111	0.094	0.125	0.097	0.127	0.088	0.098	0.079	0.077	0.006	0.004
Na	0.001	0.001	0.008	0.009	0.018	0.001	0.008	0.005	0.010	0.002	0.001	0.007	0.002
K	0.002	0.002	0.004	0.004	0.006	0.001	0.002	0.002	0.003	0.002	0.002	0.008	0.001
Ni	0.586	0.654	0.719	0.769	1.026	0.956	1.067	1.236	1.288	1.380	1.925	0.044	0.048
Total	5.076	5.090	5.043	5.041	5.042	4.632	4.944	4.937	4.513	4.525	5.007	4.971	4.940

*Total iron as FeO

[1] Samples designated as B and L are the ultramafic rocks from the vicinity of the Lilaunai and Barkotkai villages, whereas those labelled as K and Sp are from the ultramafic rocks in the Spin Obo-Kuh area (see Fig. 1).

[2] Assuming that all the analyses truly represent serpentine – a rather simplified assumption made only for the sake of broadly constraining the mineralogical characterization of the analyses.

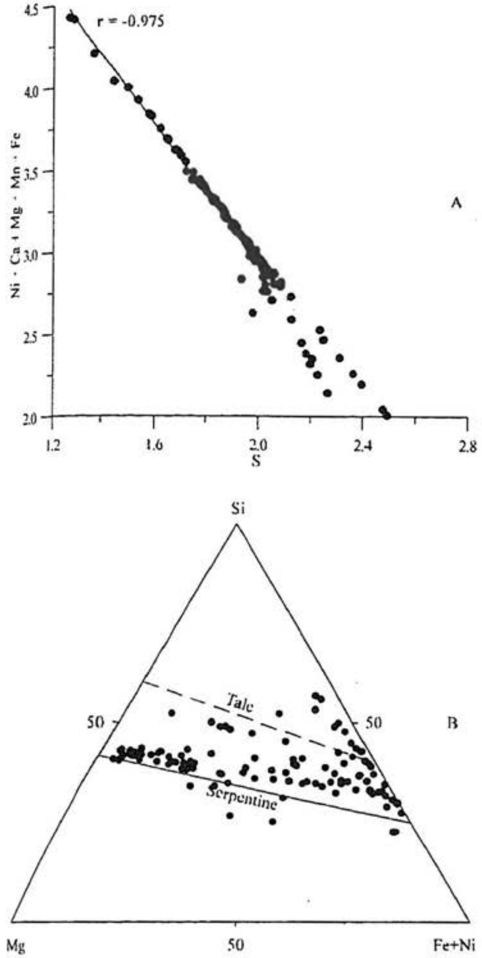

Fig. 3. Inter-element relationships and compositional variation in garnierites.

Pentlandite

Pentlandite occurs as small relic patches within the large grains of garnierites. The sizes of these relics are rather small in most occurrences, although some of the patches were large enough for analysis with a focused beam using the microprobe. Two selected analyses are given in Table 2. The S content of the analysed grains is slightly higher and, therefore, the M:S ratio (~ 1.11) somewhat lower than the stoichiometric composition of pentlandite (M:S = 1.125; see Misra & Fleet, 1973).

DISCUSSION

The distribution of the garnierite grains along fractures in the altered ultramafic rocks shows that their formation most probably commenced after the main episodes of serpentinization and carbonation of the host rocks. In addition, the presence of well preserved relics of pentlandite within the relatively large grains of garnierite strongly suggests that their formation is partly related to the alteration of original magmatic pentlandite. In other words, the garnierite formation is the result of a reaction between pentlandite and the surrounding silicates ('normal' serpentine and/or talc). This reaction, which probably took place in the presence of a fluid phase channelized by fractures, can be simplified as:

'normal' serpentine/talc + pentlandite → Ni-, Fe-rich serpentine/talc ± magnetite

TABLE 2. ANALYSES OF PENTLANDITE

No.	1	2
	Weight %	
Ni	42.40	43.75
Co	1.41	1.74
Fe	22.33	21.02
Cu	0.08	0.09
S	33.76	33.38
Total	99.97	99.98
	Atomic %	
Ni	32.84	33.97
Co	1.08	1.35
Fe	18.17	17.15
Cu	0.06	0.07
S	47.85	47.46
Metal:Sulphur	1.090	1.107

The sulphur released as a result of this reaction was most probably removed by the fluid as SO_4^{-2} ions.

The within-grain heterogeneity in the optical properties and, therefore, chemical composition of the garnierites studied is rather high and could be the result of some or all of the following factors:

1. Difference in the chemical activity of the component elements (Fe and Ni) of the precursor (pentlandite).
2. The reaction probably took place under very low temperature such that the whole of the precursor could not equilibrate/react uniformly.
3. Contrast in the physical conditions over very short distances, i.e. difference in micro-environments.

The importance of the role which the micro-environments may play in causing chemical variation has been discussed recently by Christidis and Dunham (1993) for smectites. Such a behaviour seems to hold in case of the specimens studied and probably indicates that chemical heterogeneity is a rather common characteristic of phyllosilicates formed at low temperatures.

Alternatively, the chemical heterogeneity (especially the variable and mostly high concentrations of iron and calcium) is not due to an uneven incorporation of the 'foreign' metals into the structure of the studied serpentine and/or talc granierites; rather it reflects an intimate intergrowth of garnierite with oxides and/or hydroxides of iron and carbonate (calcite). Although the probe diameter employed was <1 µm, the intergrowths might be on a much finer scale; far too fine to be noted by optical or secondary electron microscopy. Such a possibility clearly exists, as shown by the poor correlation coefficients for the Fe-Mg and Ca-Mg plots of the current analyses, and is also indicated by previous studies on similar materials from elsewhere (e.g. Hotz, 1964; Uyeda et al., 1973). However, the applicability of this possibility to the specimens investigated warrants further detailed examination by more sophisticated techniques such as analysis by transmission electron microscopy (TEM).

CONCLUSIONS

1. The dominant occurrence of garnierites along fractures in the rocks shows that their formation took place after the main episodes of serpentinization and carbonation.
2. The very high Fe content and the presence of pentlandite as relics in most of the analysed garnierite grains indicate that the latter most probably developed through a reaction between magmatic nickel sulfides and 'normal' serpentine and/or talc.
3. It appears that after serpentinization and carbonation, the ultramafic rocks suffered a further supergene alteration through Ca-rich, K-bearing carbonated fluids leading to the formation of calcite and muscovite together with garnierite. That is perhaps why the analyses of garnierites show relatively high amounts of CaO.

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