Layer silicates from Szklary (Lower Silesia): from ocean floor metamorphism to continental chemical weathering

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Abstract

The weathering crust at Szklary is known as a classical location of a nickeliferous laterite deposit derived from the chemical weathering of ultrabasic rocks. The layer silicates from the Szklary massif have been studied since the eighteenth century; moreover, this locality is considered to be an exceptional location of different minerals including nickel containing corrensite, interstratified kerolite-stevensite, interstratified serpentine-smectite, kerolite-pimelite, and clintonite. Ni-corrensite and irregularly mixed-layer serpentine-smectites with a variable layer ratio were found in Szklary for the first time.

The origin of the layer silicates from Szklary is complex: (1) serpentine, chlorite, and clintonite are products of hydrothermal metamorphism related to the serpentinization of ultramafic rocks and posterior metamorphism and (2) the mixed-layer minerals, sepiolite, and kerolite-pimelite formed due to the hydrothermal and supergeneous alteration of ultrabasic rocks and various metamorphic schists.

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INTRODUCTION

The layer silicates from Szklary and particularly their nickeliferous varieties have been a subject of interest for mineralogists for over 200 years. The first reports regarding the occurrence at Szklary of green aggregates of nickeliferous phyllosicates, presently known as garnierites, are from the eighteenth century (Klaproth, 1788 fide Lis & Sylwestrzak, 1986), and pertain to the so-called Chrysopraserde from Koźmice, a village located next to Szklary. Nickeliferous layer silicates from Szklary are present in many museum collections and are used as reference samples in mineralogical studies (e.g. Faust, 1966; Springer, 1974; Brindley et al., 1979).

For many years altered ultramafic rocks from the Szklary massif were mined as nickel ore. The saprolitic and clayey-ferruginous zones played a major economic role, as in many other lateritic nickel deposits. In these deposits, smectite (saponite, nontronite, Fe-montmorillonite, stevensite, beidellite), nickel-bearing and nickeliferous serpentines, and kerolite-pimelite series minerals were Ni-carriers (e.g. de Chételat, 1947; Besset, 1980; Golightly & Araciba, 1979; Golightly, 1981; Boukili et al., 1983; Fontanaud & Meunier, 1983; Pelletier 1983; Colin et al., 1985, 1990).

GEOLOGICAL CHARACTERISTICS

The Szklary massif is located in the Niemcza dislocation zone, east of the Góry Sowie border. It is the smallest of the three ultrabasic massifs situated around the Góry Sowie block, and it is regarded as mantle peridotites of a

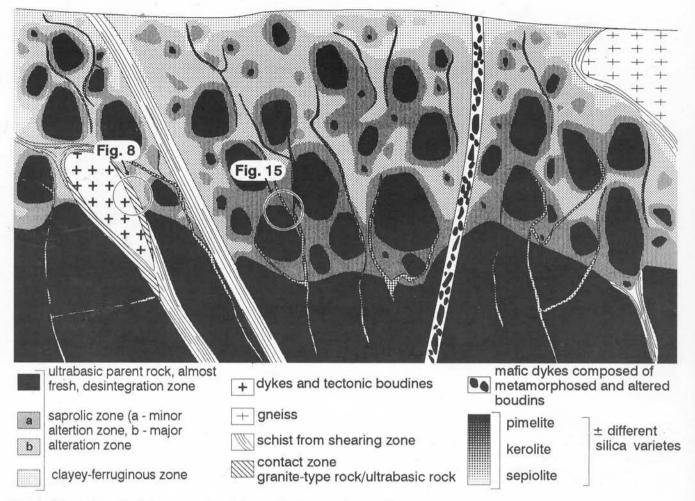


Fig. 1. Schematic geological cross-section of the weathering crust from Szklary.

tectonically dismembered Palaeozoic ophiolitic sequence (Pin et al., 1988). Comparing the ultramafic of the Jordanów-Gogołów and Braszowice-Brzeźnica massifs which typically have normative harzburgite composition to rocks from the Szklary massif rocks, the latter show more variable modal and normative compositions mainly in lherzolites, websterites and pyroxenites (Szymkowiak, 1981; Dubińska & Gunia, 1997). The basic geochemical characteristics of the Szklary massif ultramafic rocks are evidence of their variability; besides residual mantle (MORB-type) rocks there are rocks with a geochemical signature suggesting enriched residual mantle (ERM, Dubińska & Gunia, 1997). The ultramafic rocks from the Szklary massif are generally highly overgrown with monoclinic amphibole, and to a lesser degree with orthorhombic amphibole. Besides the ultrabasic rocks, it is possible to find small leucocratic granite-type bodies, tectonically embedded gneiss fragments, probably derived from the Niemcza dislocation zone as well as boudines and boudined dykes of mafic rocks including metarodingites, amphibolites and mafic granulites (720-760°C, 3.5 kbar), displaying boninitic affinity (Dubińska et al., 1991; Gunia, 1995; Dubińska, 1997; Dubińska & Gunia, 1997).

The rocks of the Szklary massif are highly altered, analogically to other weathering crusts from the Sudety Mts. These alterations are attributed to Tertiary chemical

weathering. The structure of the Szklary weathering crust is typical for a New Caledonian laterite, i.e. in which fissures, usually of tectonic origin, played a major role as channels for percolating solutions (e.g. de Chételat, 1947; Trescazes, 1973; Besset, 1980; Nahon et al., 1982). Other pieces of evidence for such a genetic model are the variable thickness of the weathered zone, sometimes up to 100 m (after Birecki et al., 1962; Niśkiewicz, 1967; Niśkiewicz et al., 1979), and its mosaic structure. Compared to the thickness of other known weathering crusts developed on ultrabasic massifs, the crust at Szklary (originally up to 100 m thick) is very unusual, for example the thickness considered standard for New Caledonian weathering crust on ultramafic rocks is generally ca. 50 m (Troly et al., 1979), seldom reaching 80 m (Orloff, 1968 vide Besset, 1980). In most cases the thickness of this type of crust does not exceed 30 m (e.g., Zeissink, 1969; Ogura et al., 1986; Colin et al., 1990; Martini 1994). Although the Szklary crust is considered to be exceptionally thick, neither ferruginous cuirasse, (a zone composed of SiO2 varieties and iron oxides, oxyhydroxides and hydrous oxides), nor an upper horizon of laterite developed at the expense of ultrabasic rock were reported. The above-mentioned zones may be Al2O3-enriched to a degree so they might contain kaolinite and gibbsite (e.g., Nikitina et al., 1971; Paquet et al., 1982; Boukili et al., 1983; Colin et al., 1990; Das et al.,

1999). The lack of those horizons proves that the original thickness of the weathered profile at Szklary most likely exceeded 100 m. The complex structure of this weathered crust is a result of the diversification of the altered material including: ultrabasic rocks, a variety of acid crystalline rocks – granitic rocks (Variscan?), gneisses, and basic rocks – amphibolites, rodingites and products of their metamorphism. Figure 1 shows a schematic sketch of the weathered crust at Szklary.

The complex geological evolution of the Szklary massif is reflected in its exceptional variety of layer silicate assemblages. A small area of about 10 km² also includes endogenic layer silicates, products of hypergenic alteration of ultrabasic rocks, amphibole-chlorite schist, granite-type and mafic dykes, and gneisses from the Niemcza dislocation zone directly bordering the ultrabasic rocks of Szklary massif. The extensive quantity of phyllosilicates found here comprises the following groups of minerals:

- serpentine group minerals (antigorite, lizardite and chrysotile) - products of low temperature serpentinization of ultramafic rocks and their posterior metamor-

phism;

- clintonite, common in the metarodingites at Szklary (rodingite - calc-silicate rocks, SiO₂-undersaturated rock formed as a by-product of serpentinization; e.g., Honnorez & Kirst, 1975; O'Hanley et al., 1992; O'Hanley, 1996; Dubińska et al., 1988, 1991; Dubińska, 1997);

 magnesium chlorites, common in most ultrabasic and basic rocks, shearing zones, and contact zones surrounding apophyses of leucocratic rocks (Ostrowicki, 1965a; Wiewióra, 1978; Wiewióra & Dubińska, 1987);

– kerolite-pimelite (talc-like) minerals; we have also included interstratified kerolite-stevensite, which is genetically related to kerolite-pimelite, in this group of minerals (Wiewióra, 1959; Ostrowicki, 1965a; Wiewióra et al., 1982; Dubińska, 1984; Sakharov et al., 1998a);

- talc, common in most of the Szklary rocks; massive talc aggregates occur in contact zones surrounding granite-type dykes (Ostrowicki, 1965a; Dubińska, 1982a);

- interstratified serpentine-smectite and serpentinevermiculite; this type of mixed-layer minerals has not yet

been reported as a natural occurrences;

- smectites: (1) a nickel-bearing ferrous analogue of montmorillonite, a common product of ultrabasic rock alteration in saprolitic zones (Dubińska, 1986); (2) montmorillonite-beidellite, a typically result of mafic dyke altera-

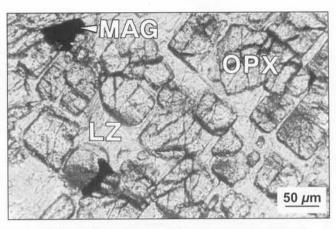


Fig. 2. Ultramafic rock with relict orthopyroxene (OPX) intergrowth with lizardite (LZ) flakes; containing magnetite (MAG) formed at the expense of the primary interstitial chromite, sample WK3 (courtesy of Andrzej Szymkowiak), one polar.

tion (Ostrowicki, 1965a); (3) trioctahedral smectite (saponite?) usually formed at the expense of tremolite (Dubińska, 1981, 1982a);

- kaolinite group minerals (kaolinite, rare halloisite), common products of the chemical weathering of granite-type rocks and small bodies of gneiss, tectonically displaced into the Szklary ultrabasic rocks (Ostrowicki, 1965b; Dubińska, 1982a);

 sepiolite occurring in veins in somewhat weathered ultrabasic rocks (Ostrowicki, 1965b; Dubińska, 1982a);

- interstratified minerals containing chloritic layers, including regularly mixed-layer trioctahedral chloritesmectite (corrensite, Wiewióra & Szpila 1975; Wiewióra & Anulewicz, 1976).

The altered ultrabasic rocks from Szklary, particularly in varieties containing kerolite-pimelite, typically consist of different aggregates (pockets, veins, veinlets) of silica minerals: chalcedony, CT-opal, microcrystalline-and megaquartz. Mixed-layer serpentine-smectite can also occur with SiO₂-group minerals.

Most of the layer silicate groups described below are economic nickel carriers in lateritic ores and can also be

used as geological markers.

SERPENTINES

The primary ultramafic rocks of the Szklary massif, including lherzolite, harzburgite, wehrlite, clinopyroxenite (Spangenberg & Müller, 1949; Juskowiak, 1957; Niśkiewicz, 1967, 1993; Szymkowiak, 1981; Dubińska & Gunia, 1997) are frequently intergrown with serpentines. The complexity of the Szklary serpentinites is resulted of parent periods of tectonic movements, along with the continuing formation of serpentine group minerals. Pseudo-

morphic varieties of serpentinite, typically rich in lizardite (e.g., Wicks & O'Hanley, 1988; O'Hanley, 1996), are scarce in Szklary. They formed at the expense of rocks with abundant olivine, which was intergrown with one or more generations of monoclinic amphibole. In general, small proportions of the relict minerals are serpentinized; the olivine is frequently untouched in rocks without evident serpentinization (Szymkowiak, 1981). Pyroxenites are typically partially intergrown with lizardite (Fig. 2)

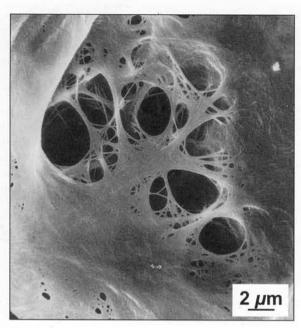


Fig. 3. Chrysotile (?) fibers in sepiolite-kerolite rock, sample 66, SEM, photo Andrzej Rochewicz

and/or antigorite and they are overprinted to various extents with monoclinic amphiboles. The ultramafic rocks of Szklary contain numerous cross-fiber veins (chrysotile asbestos). Although serpentine group minerals are typically completely obscured by various weathering products, residual chrysotile fibers can be found in kerolite-sepiolite veinlets (Fig. 3).

CLINTONITE

Clintonite is trioctahedral brittle mica containing Ca²⁺ ions in the interlayer. Clintonite is one of the CaAlsilicates present in the ophiolitic calc-silicate rocks, which were produced by greenschist facies metamorphism (Rice, 1983; O'Hanley, 1996) that overlapped onto early products of Ca-metasomatism (rodingites) formed simultaneously with serpentinization. At Szklary, clintonite occurs in metarodingite and it is associated with epidote, grossularite-rich garnet, diopside, spinel close to the Mg2AlO4 end-member (Figs. 4 and 5) and monoclinic amphibole (Dubińska, 1997). Fluid-inclusion data suggest that the formation temperatures of garnet, epidote, and clintonite overlapped (300-370°C depending on the assumed ordering of the epidote structure), whereas the diopside was produced by a higher temperature episode, 400°C, which is consistent with textural observations (Dubińska et al., 1991); both suggest greenschist facies metamorphism associated with antigorite formation in the ultrabasic rocks. The common occurrence of clintonite-bearing metarodingites in the Szklary massif implies that the extent of zeolite facies metamorphism (ocean floor metamorphism?) was more widespread than could be considered on basis of the limited occurrence of the pseudomorphic variety of serpentinites at Szklary.

Table 1
Representative microprobe analyses of clintonite from rodingites

				8			
sam ple no.	Sz100A	Sz100A	Sz100A	Sz100A	Sz90B	Sz90B	Sz90E
SiO ₂	15.81	15.48	16.30	15.30	16.69	17.38	16.69
TiO_2	0.05	0.10	0.06	0.03	- 2	0.01	0.02
Al_2O_3	40.97	39.88	38.76	40.17	41.76	41.82	41.17
Cr ₂ O ₃	n.d.	0.11	0.05	0.12	0.14	0.09	0.22
Fe ₂ O ₃ tot	2.44	1.87	1.87	1.96	3.76	3.53	3.44
MnO	0.11	0.04		iz.		0.01	+
NiO	0.42	0.22	0.25	0.30	0.32	n.d.	0.46
MgO	18.94	18.70	18.90	18.58	18.31	19.26	18.05
CaO	12.69	12.33	12.18	12.35	13.03	13.01	12.8
K ₂ O	- 0.03		0.07	0.02	0.03		0.04
Na ₂ O	0.03	0.02	0.12		0.08	0.18	0.03
total	91.47	88.78	88.56	88.83	94.12	95.29	92.92
		on	the basis	of O10(O	H) ₂		
Si	1.17	1.17	1.24	1.16	1.20	1.23	1.21
Ti		0.01					
Al	3.56	3.56	3.47	3.59	3.53	3.48	3.53
Cr		0.01		0.01	0.01	0.01	0.01
Fe ³⁺	0.14	0.11	0.11	0.11	0.20	0.19	0.19
Mn	0.01						
Ni	0.02	0.01	0.02	0.02	0.02		0.03
Mg	2.08	2.11	2.14	2.10	1.96	2.03	1.95
Ca	1.00	1.00	0.99	1.00	1.00	0.99	1.00
K			0.01				
Na			0.02		0.01	0.02	
Al^{IV}	2.83	2.83	2.76	2.84	2.8	2.77	2.79
Al^{vi}	0.73	0.73	0.71	0.75	0.73	0.71	0.74
Σ octa- hedral cat.	2.98	2.97	2.98	2.98	2.91	2.93	2.91

- not detected; n.d. - not determined

Apparently, the symptoms of very low-grade metamorphism of the ultrabasic rocks are obscured due to Variscan metamorphic recrystallization and Tertiary chemical weathering.

The composition of the clintonite from Szklary is close to the theoretical formula Ca(Mg,Fe,Al)₃(Al,Fe³⁺,Si)₄O₁₀(OH)₂.

The Al^{IV} contents, frequently more than 2 (on the basis of T₄O₁₀(OH)₂, see Table 1), is analogous to clintonites from other occurrences (e.g. Harada *et al.*, 1965; Forman *et al.*, 1967; MacKinney *et al.*, 1988). The chemical composition of the Szklary clintonite is quite monotonous (Fig. 6) indicating the mineral developed within one episode and that posterior geological events did not modify its chemistry.

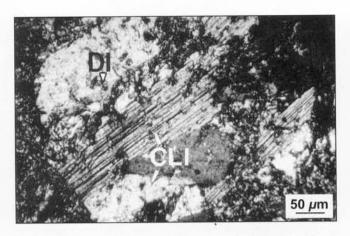


Fig. 4. Clintonite (CLI) intergrown with newly formed diopsidic pyroxene (DI) in rodingite, sample 739, crossed polars.

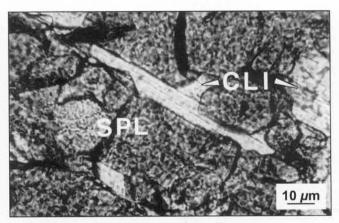


Fig. 5. Clintonite (CLI) flakes intergrown with spinel (SPL) in rodingite, sample Sz93, one polar.

CHLORITES

Chlorites are ubiquitous minerals in many rocks of the Szklary massif. Most of the ultrabasic rocks are intergrown with Mg-chlorite. Among the shear zone rocks it is typically find amphibolite-chlorite schists and occasional almost monomineral chlorite schists. Chlorite schists also occur in the highly tectonized contact zones surrounding minuscule leucocratic apophyses (Fig. 8), where they accompany other schists containing minerals having preliminary X-ray characteristics similar to vermiculite (Ostrowicki, 1965a; Wiewióra, 1978; Dubińska 1982a, b; Wiewióra & Dubińska, 1987).

The size of the chlorite flakes often exceeds 0.5 cm. Minor quantities of chlorite also exist in the mafic rocks – amphibolite and rodingite. Regardless of the rock type containing the chlorite from Szklary, it is chemically close to clinochlore (the Mg end-member of the clinochlore-chamosite series) although the concentration of aluminium and iron varies significantly, particularly in the chlorites from the rodingites (Fig. 6, Table 2). The nickel concentration in the chlorites is variable, however it does not exceed 1% wt. of NiO (Table 2). The chlorites from Szklary represent the polytype IIb (according to Bailey, 1980), e.g. Wiewióra (1978) and Wiewióra & Dubińska (1987).

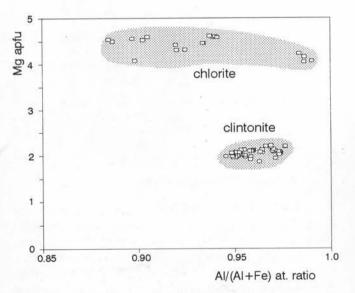


Fig. 6. Diagram showing variations in the chemical compositions of the clintonite and chlorite from rodingites; apfu – atoms per formulae unit, at. ratio – atomic ratio; microprobe analyses calculated on the basis of O10(OH)8 and O10(OH)2 for chlorite and clintonite, respectively.



Fig. 7. Chlorite schist from a shear zone, sample 69, crossed polars.

MIXED-LAYER MINERAL SERPENTINE-SMECTITE AND SERPENTINE-VERMICULITE

The thus-far identified interstratified minerals with serpentine layers contain serpentine and chlorite layers (e.g., Bons & Schryvers, 1989; Jiang et al., 1992; Slack et al., 1992; Bailey et al., 1995; Banfield & Bailey, 1996; Dalla Torre et al., 1996; Ryan & Reynolds, 1996; Xu et al., 1996, Schmidt et al., 1999). According to the authors of this text only one case of synthetic interstratified structure lizardite-smectite has been reported so far (Torii et al., 1998; Nagase et al. 2000).

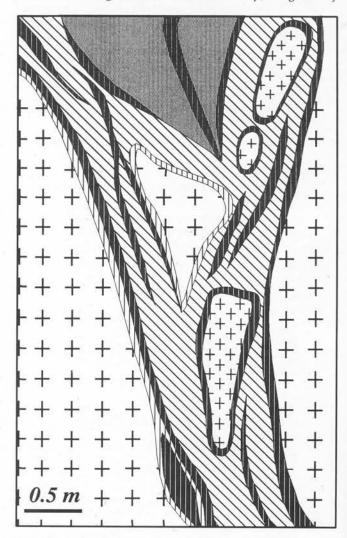
At Szklary interstratified minerals with serpentine

Table 2
Representative analyses of chlorite from rodingites and amphibolites

rock type	schist from shear- ing zone	5-	rodingi	te	amphibolite					
sample no.	Sz69(1)	739	Sz93A ¹	Sz100A	Sz9A	Sz90A	Sz90A	Sz90A		
SiO ₂	31.53	28.07	30.88	34.55	28.21	28.39	28.05	29.01		
TiO ₂	0.01	n.d.		0.01	*			0.03		
Al_2O_3	17.46	22.73	20.47	19.51	22.43	24.58	22.92	21.12		
Cr ₂ O ₃	0.12	n.d.	0.03	- 2	0.05	-	0.32	17.		
FeO tot.	4.91	2.00	3.76	0.42	5.52	2.44	3.70	4.06		
MnO	0.02	n.d.	0.08	- 2	0.05	0.04	0.03	0.08		
NiO	0.19	n.d.	n.d.	0.28	0.12	n.d.	0.76	0.26		
MgO	32.81	32.00	32.55	31.61	28.56	32.04	29.28	29.73		
CaO	-	-	0.09	0.02	0.08	0.01	7.0	0.02		
K ₂ O	0.04	25	121			12				
Na ₂ O	0.07	0.04	0.01		8		0.01			
total	87.15	84.84	87.87	86.40	85.02	87.50	85.07	84.31		
			on the b	asis of O	10(OH)					
Si	3.00	2.70	2.89	3.20	2.75	2.65	2.72	2.83		
Al	1.96	2.57	2.26	2.13	2.58	2.70	2.62	2.43		
Cr							0.02			
Fe ²⁺ tot.	0.353	0.16	0.29	0.03	0.45	0.19	0.30	0.33		
Mn			0.01					0.01		
Ni	0.02			0.02	0.01		0.06	0.02		
Mg	4.65	4.58	4.54	4.36	4.15	4.45	4.23	4.33		
Al ^{IV}	1.00^{3}	1.30	1.11	0.80	1.25	1.35-	1.28	1.17		
$A1^{VI}$	0.963	1.27	1.15	1.33	1.33	1.35	1.34	1.26		
Σ octa- hedral cat.	5.98	6.01	5.99	5.74	5.94	5.99	5.93	5.95		
Mg/ (Mg+ Fe)²	0.93	0.97	0.94	0.99	0.90	0.96	0.93	0.93		

1. inclusion in clintonite; 2. atomic ratios; 3. structural formulae of chlorite SZ69(1): $(Mg_{4.65}Al_{1.00}Fe^{2+}_{0.28}Ni_{0.02})^{VI}(Si_{3.00}Al_{0.96}Fe^{3+}_{0.04})^{IV}O_{10}(OH)_8$, determinations of Fe^{3+VI} , Fe^{2+VI} , and Fe^{3+IV} on the basis of Mössbauer spectra, unpublished data; n.d. – not determined; – – not detected

layers were found in the altered contact zones between ultrabasic and leucocratic rocks (Fig. 8). Analogous minerals occur in the altered biotite schists at Wiry (western part of the Jordanów-Gogołów serpentinite massif, Sakharov et al., 1998a). Both at Szklary and at Wiry, mixed-layer minerals containing serpentine layers are late products of the vermiculitization of trioctahedral mica from the contact schists. Rocks from the Jordanów-Gogołów serpentinite massif, including contact schists from Wiry, are generally



altered ultrabasic rock

altered biotite schist

chlorite schist

+ altered granite-type rock

+ altered aplite

tremolite schist

Fig. 8. Sketch of the occurrence of contact zones between ultrabasic and granite-type rocks.

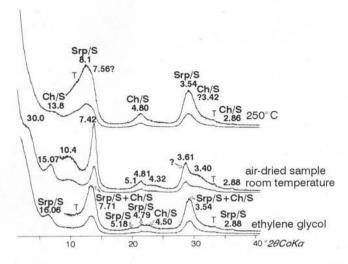


Fig. 9. Experimental X-ray patterns of sample Sz50 (ϕ < 0.5 μ m), oriented aggregates; 250°C – X-ray pattern recorded without cooling the specimen; Ch/S – interstratified chlorite(70%)/saponite(30%), chlorite with heavy cations (Ni²+, Fe²+) preferentially located in the incomplete brucite-like sheet; Srp/S – interstratified serpentine(80%)/smectite(20%); T – talc.

not weathered, consequently the start of interstratified serpentine-smectite formation should be considered earlier that the intense Tertiary weathering (Jelitto *et al.*, 1993; Dubińska *et al.*, 1995; Bylina, 1996).

A content of serpentine and expandable (smectite or vermiculite) layers in irregularly mixed-layer serpentine-expandables, where R (Reichweite) equals zero and denotes junction probability, is different. The serpentine/expandables ratio depends on the grain fraction. Fine-particle grain fractions (ϕ <0.5 μ m) typically contain interstratified serpentine-expandable minerals, including major serpentine layers (ca. 80%), where the swelling layers can be both vermiculite and stevensite-type. The mixed-layer mineral from coarse grain particles (ϕ >5 μ m) contain predominant vermiculite layers (up to 90% in the interstratification).

The influence of disordered mixed-layer serpentineexpandables on diffraction maxima intensities is minor, hence an admixture of ordered phases can mask their presence. This is probably the reason why there were problems with the proper identification of this mineral group, particularly in the case of the overlapping of the diffrac-

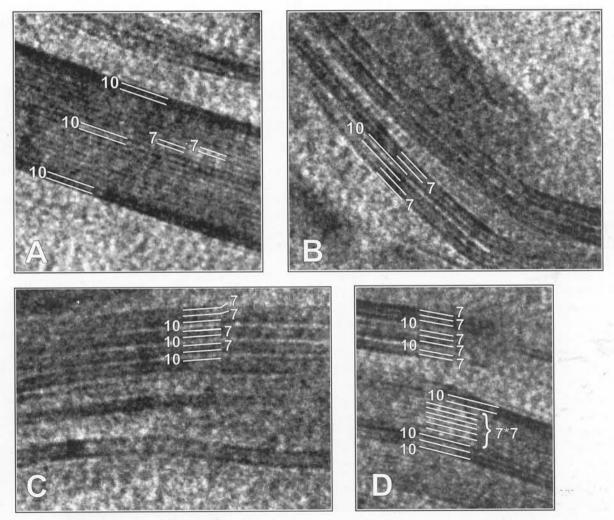


Fig. 10. HRTEM image of the interstratified serpentine-smectite domains; A. serpentine rich domain with singular smectite layers, B. five-layer domains containing a sequence 7-7-10-7-7, C. quasi-ordered domain composed of serpentine and smectite, D. domains with different sequences of serpentine and smectite layers; an ultramicrotome section of an oriented specimen cut perpendicularly to (00l); 7 – serpentine layer (7Å), 10 – contracted smectite layer (10Å).

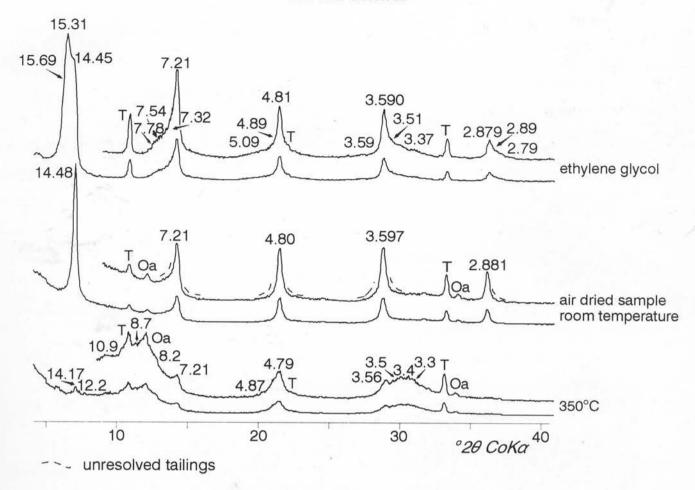


Fig. 11. Experimental X-ray patterns of sample Sz36 (2–5 μ m), oriented aggregates; 350°C – X-ray pattern recorded without cooling the specimen; Oa – orthorombic amphibole, T – talc.

tion maxima of the interstratified phase and reflections of other components including clay minerals.

The identification of mixed-layer minerals containing swelling and serpentine layers was possible by a trial and error process. This method was used until the best fit between calculated and experimental curves for ethylene glycol solvation, heated and air-dried sample treatments was achieved as described by Sakharov et al. (1999). Figure 9 shows example X-ray tracings for a sample rich in interstratified mineral serpentine-smectite. The sample contains 95% interstratified serpentine (80%) with smectite (20%) and a minor admixture, 5% disordered mixed-layer chlorite (70%) with smectite (30%); in the mixed-layer chlorite-smectite nickel is preferentially located in a brucite-like sheet (Sakharov et al., 1998a). The ASN program based on the mathematical formalism of Drits and Sakharov (1976) was used to simulate XRD (X-ray diffraction) diagrams for the interstratified minerals.

The HRTEM (High-Resolution Transmission Electron Microscopy) results correspond closely with the structural model of the simulations derived from XRD analysis, i.e. serpentine-smectite interstratification. HRTEM images show zones composed of various serpentine/smectite packets with different serpentine and smectite layer ratios: 3:1, 2:1, 1:1, 4:1, as well as scarce serpentine

tine segregations (Fig. 10). The apparent discrepancy between the XRD and HRTEM investigations is a consequence of statistical information obtained from the powder XRD study and direct observations of individual domains using the electron microscope.

Figure 11 shows that the X-ray pattern for the airdried sample is similar to vermiculite. By trial and error, a good correlation between calculated and experimental XRD patterns was obtained for different treatments (Fig. 12) revealing the following interstratified minerals in this sample:

- serpentine (80%)-vermiculite (20%), R = 0;

 low charge (LC) vermiculite (50%)-high charge (HC) vermiculite (25%)-chlorite (25%);

- LC vermiculite (30%)-HC vermiculite (40%)-chlorite (30%);

- chlorite (25%)-degraded chlorite with incomplete brucite-like sheet (60%)-HC vermiculite (15%).

The chemical composition of particles of different grain-size (Table 3) shows that the fine-grained particles are rich in mixed-layer serpentine-smectite, as evidenced by the (Mg+Ni+Fe)/Al ratio increasing with particle size reduction (Fig. 13). Nickel distribution among the different grain fractions shows an analogous trend, suggesting interstratified serpentine-smectite to be an important Ni-

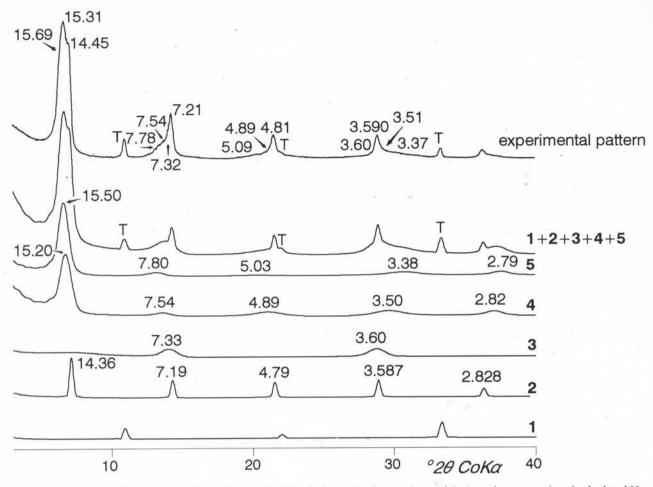


Fig. 12. X-ray tracings of sample Sz36 (2–5 μ m) treated with ethylene glycol; experimental (oriented aggregates) and calculated X-ray patterns (1, 2, 3, 4, 5) of constituent layer silicates: 1 – talc (T); 2 – Ch1 14.3Å/Ch2 $^{\circ}$ 14.4Å/V $^{\circ}$ 14.2Å = 25%/60%/15%, csd = 12–25; 3 – V $^{\circ}$ 14.2Å/Srp 7.25Å = 20%/80%, csd = 4–10; 4 – V $^{\circ}$ 14.2Å/V $^{\circ}$ 16.3Å/Ch 14.3Å = 30%/40%/30%, csd = 3–9; 5 – V $^{\circ}$ 16.3 Å/V $^{\circ}$ 14.2Å/Ch 14.3Å = 50%/25%/25%, csd = 3–10; Ch – chlorite, Srp – serpentine, V – vermiculite, $^{\circ}$ – one layer complex with ethylene glycol, $^{\circ}$ – two layer complex with ethylene glycol, csd – number of coherently scattering domains.

carrier, while the aluminium content is lowest the in finegrained particles ($\phi < 0.5 \mu m$).

The formation of an interstratified mineral assemblage does not influence the texture of the parent schist; its schistosity is still distinguishable, with narrow idio-

morphic quartz grains in between phyllosilicates flakes (Fig. 14) and inhomogeneous sceptre-shaped grains of large quartz with zones of later small quartz enclosing their prism faces.

KEROLITE-PIMELITE SERIES (INCLUDING MIXED-LAYER KEROLITE-STEVENSITE)

The kerolite-pimelite series is a fine-grained and turbostratic analogue of talc-willemseite, where talc and kerolite represent magnesium end-members, while pimelite and willemseite are their nickel analogues. At Szklary, kerolite-pimelite occurs (1) in a network of fine veinlets and fissure fillings in ultrabasic rocks, chlorite-, amphibole-chlorite- and amphibole schist, typically more or less altered and (2) in veins as a continuation of sepiolite-bearing veins (Fig. 15). In the fine veinlets (1) kerolite-pimelite minerals are concomitant with quartz and CT-opal. The mineral assemblages of the vein (2) are diversified and related to the weathering rate of the adjacent rock. The vein mineralogy changes gradually (Fig. 15). Veins penetrating almost fresh rocks are composed of major sepiolite with an admixture of kerolite, and/or magnesite, magnesium calcite, serpentine, and quartz. Highly weathered rusty rocks from the clayey-ferruginous zone contain veins consisting of kerolite-pimelite, typically with CT-opal and quartz. According to Gajewski (1970) this type of vein can occur in the Szklary massif to a depth of 120 m.

The kerolite-pimelite aggregates from the veins usu-

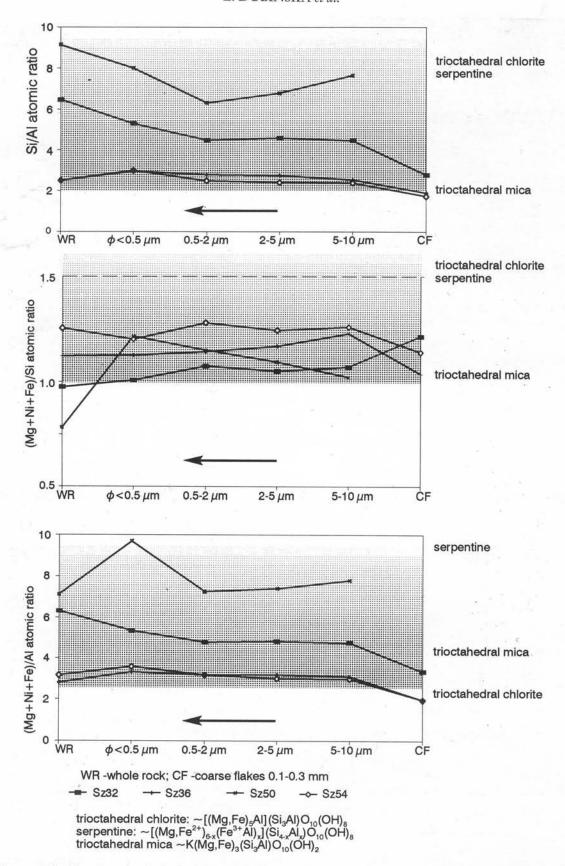


Fig. 13. Diagrams showing the chemical diversity of different grain fraction samples containing interstratified serpentine-smectite(vermiculite); samples represent altered contact zones from between ultrabasic and granite-type rocks. High ratios of (Mg+Ni+Fe)/Si, Mg+Ni+Fe)/Al, and Si/Al in the fine-grained material $(\phi < 0.5 \mu m)$ compared to the whole rock and coarse grain fraction compositions are related to increasing mixed-layer serpentine-smectite(vermiculite) content in the former; arrows show the rising concentration of interstratified serpentine-smectite(vermiculite). Sample Sz50 does not contain coarse flakes; moreover, its grain fraction 5–10 μ m is quartz-rich.

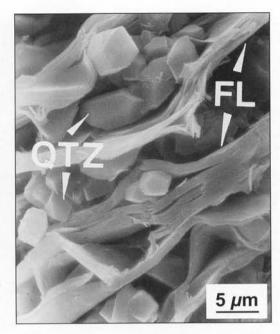


Fig. 14. Newly formed quartz (QTZ) intergrowth in altered biotite schist, FL - flakes of layer silicates, sample 8, SEM, photo Andrzej Rochewicz.

ally represent fine-grained flaky clusters (Fig. 16) but seldom fibrous clusters (Dubińska, 1984). Homogeneous films replaced by honeycomb coatings of kerolite-pimelite can occur in the slightly weathered ultrabasic rocks adjacent to the veins (Figs. 17 and 18).

The X-ray characteristics of most of the kerolitepimelite series from Szklary (Figs. 19 and 20) are close to those of kerolite-pimelite from other occurrences (e.g. Maksimović, 1966; Brindley et al., 1979). The layer stacking of the kerolite-pimelite from Szklary is turbostratic (disordered) as evidenced by broad two-dimensional hk bands (Fig. 19). The kerolite-pimelite partially expands after ethylene glycol treatment (Fig. 20); this swelling can result from a segregation of expanding, i.e. stevensite-like, and non-expanding domains. However, this expansion is frequently attributed to weak van der Waals bonds between talc-type layers (Brindley et al., 1977, 1979). That the Szklary kerolite shows an increase of expansion after ultrasonic treatment (Wiewióra et al., 1982) supports Brindley's (op. cit.) suggestion. The ultrasonic treatment produced a regularly interstratified (R=1) structure containing talc and stevensite layers, 75% and 25% respectively, with coherently scattering domains (csd) comprising 2-6 layers (lognormal distribution), while the parent kerolite XRD patterns were typical for the segregation model of kerolite structure (Sakharov et al., 1998a).

An unusual interstratified kerolite-stevensite occur-

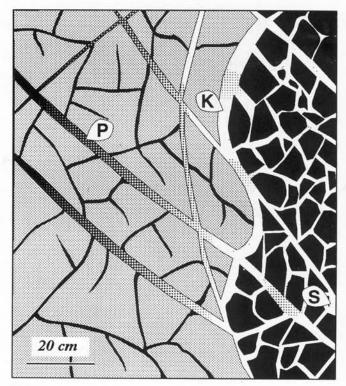
Table 3
Chemical compositions of grain fractions and coarse flakes of the altered biotite schist (on anhydrous basis)

	SiO ₂	TiO ₂	Al_2O_3	Cr ₂ O ₃	FeO ¹	NiO	MnO	MgO	CaO	Na ₂ O	K_2O	Mg/Ni3
sample Sz32												
φ<0.5 μm	48.27	0.13	7.72	0.12	5.02	18.91	0.05	19.03	0.61	0.02	0.12	1.77
0.5-2 μm	46.89	0.12	8.90	0.12	4.68	16.22	0.05	21.93	0.96	0.02	0.11	2.38
2-5 μm	47.46	0.11	8.78	0.13	4.56	14.34	0.06	22.76	1.70	0.03	0.07	2.79
5-10 μm	47.16	0.11	8.96	0.12	4.39	13.67	0.06	23.56	1.86	0.04	0.07	3.03
32 flakes²	44.13	0.25	13.58	0.09	4.11	8.90	0.02	28.57	0.35			5.65
sample Sz36												
φ<0.5 μm	45.91	0.38	13.17	0.16	6.40	4.13	0.06	28.70	0.68	0.05	0.36	12.23
0.5-2 μm	45.24	0.79	13.66	0.16	7.67	2.35	0.07	29.01	0.67	0.02	0.36	21.66
2-5 μm	44.63	0.71	13.82	0.15	7.67	2.94	0.08	29.00	0.74	0.02	0.24	17.33
5-10 μm	43.16	0.64	14.51	0.16	7.84	2.96	0.09	29.49	0.96	0.02	0.17	17.53
36 flakes²	43.25	0.96	19.44	0.16	7.94	2.97	0.09	23.94	1.21		0.04	14.18
sample Sz50											19-00	
φ<0.5 μm	46.67	0.49	4.95	0.15	3.80	18.39	0.02	25.40	0.06	0.01	0.06	2.43
0.5-2 μm	48.19	0.06	6.45	0.19	4.52	13.42	0.06	26.96	0.03	0.06	0.06	3.54
2-5 μm	48.90	0.06	5.99	0.19	4.96	11.31	0.08	28.26	0.17	0.02	0.06	4.01
5-10 μm	51.52	0.06	5.72	0.21	5.26	10.25	0.10	26.55	0.25	0.02	0.06	4.56
sample Sz54												
φ<0.5 μm	44.44	0.36	12.59	0.16	6.37	7.07	0.06	28.19	0.68	0.06	0.02	7.00
0.5-2 μm	42.73	0.01	14.59	0.15	5.05	6.76	0.04	29.99	0.58	0.05	0.05	7.81
2-5 μm	43.23	0.01	15.20	0.16	5.08	5.64	0.03	30.03	0.56	0.02	0.04	9.36
5-10 μm	42.74	0.01	15.33	0.16	4.99	6.86	0.03	29.37	0.48	0.01	0.02	7.54
54 flakes ²	42.00	0.20	20.76	0.09	4.50	5.90	0.01	26.21	0.33		-	7.82

¹⁻total iron content as FeO, 2-coarse grain fractions (0.1-0.3 mm, obtained by hand picking under binocular microscope, 3- atomic ratio

rence was also reported from the weathering crust at Szklary. The exceptional expansion of this mineral is noted at different relative humidities (R.H.), particularly in the 90-100% range, as shown in Figure 21. This mineral was identified using the best fit technique of calculated and experimental X-ray curves for a magnesium saturated sample, followed by heating and ethylene glycol treatment. After glycol treatment the mineral showed a three component mixed-layer structure K (62%)-S1 (30%)-S2 (8%), where K denotes non-expanding layers (9.6 Å, kerolite), S1 - stevensite with two layers of ethylene glycol (17.1 Å), and S2 - stevensite with one layer of ethylene glycol (13.2 Å), R=1, csd 2-5 (lognormal distribution). Both types of stevensite layers contracted to 9.6 Å after heating (Sakharov et al., 1998b). The best agreement of simulated and experimental XRD patterns was achieved for defective kerolite structure with discontinuous tetrahedral sheets (7% empty space) analogous to the model of hydrated talc described by Drits and Tchoubar (1990).

Table 4 shows the chemical composition of the kerolite-pimelite minerals and mixed-layer kerolite-stevensite from Szklary. The original analyses were corrected taking into consideration admixtures of quartz, serpentine and chlorite (see Figs. 19–21). The nickel concentration in the kerolite-pimelite minerals may be 100 times greater than those in the altered ultramafic rocks from Szklary. Kerolite-pimelite is a very frequent component of the Szklary weathering crust, thus it is an important nickel carrier. An admixture of Ni-rich kerolite-pimelite gives a green colouration to the well-known Szklary chrysoprase (Sachanbiński, 1980).



ultrabasic rock, slightly altered

highly altered ultrabasic rock from clayeyferruginous zone

veinletes containing pimelite (P) - kerolite (K) - sepiolite (S) ± admixture of silica varietes

Fig. 15. Sketch of the occurrence of sepiolite-kerolite-pimelite veins in variously altered ultrabasic rocks.

SEPIOLITE

Sepiolite commonly occurs in veins filling the fissures of slightly altered ultrabasic rocks, frequently in the presence of kerolite-pimelite (Fig. 15). The consistency of fresh sepiolite-rich samples is similar to thick cream, while dried samples are like meerschaum. SEM (Scanning Electron Microscopy) image indicates that the sepiolite aggregates look like hypha (Fig. 22), suggesting direct mineral crystallization from a solution. Diffractograms of the sepiolite-bearing samples are shown in Figures 23 and 24. The expansion of the studied sepiolites saturated with eth-

ylene glycol documents their structural disorder (see Güven and Carney, 1979).

The origin of the sepiolite from Szklary is ambiguous. Sepiolite is often reported in weathering crusts as well as in hydrothermally altered rocks (Jones & Galan, 1988). The Ni concentration in the sepiolite-kerolite veins sometimes exceeds 20% wt. (as NiO). The chemical composition of the sepiolite from Szklary is close to magnesian end-member, thus kerolite-pimelite minerals are the Nicarriers (Dubińska, 1984).

INTERSTRATIFIED MINERALS WITH CHLORITE LAYERS INCLUDING CORRENSITE

Interstratified minerals with chlorite layers were found in the altered contact zones developed between the leucocratic and ultrabasic rocks and in the altered chlorite schist from the shear zones (Dubińska, 1982b). Highly altered and weathered samples, frequently soft and plastic,

still preserve their parent schistosity.

The X-ray diffraction study revealed that mixed-layer phases containing chlorite layers represent irregular interstratification (R = 0) composed of chlorite and expandable (vermiculite or smectite) layers with chlorite/expandable

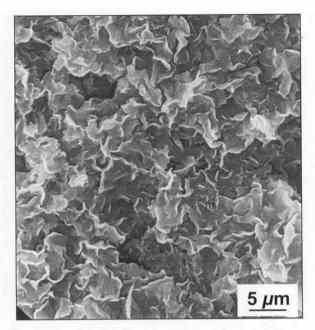


Fig. 16. Platy grains of kerolite-pimelite from a veinlet in altered ultrabasic rock, sample 74, SEM, photo Andrzej Rochewicz.

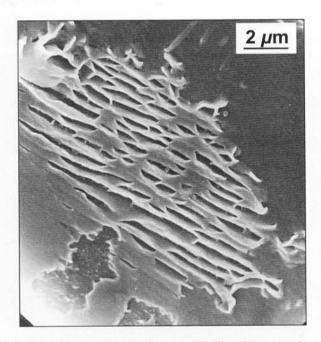


Fig. 17. Thin coating of the interstratified kerolite-stevensite on an amphibole-chlorite schist, sample 11a adjacent to sample 11, SEM, photo Andrzej Rochewicz.

ratios of 1:1 and/or 1:2 (Figs. 9, 11, and 12). The distribution of heavy cations (Ni²⁺, Fe²⁺, and Fe³⁺) is asymmetric, with their preferential location in the brucite-like sheet. The mixed-layer chlorite-expandable usually occurs together with interstratified serpentine-smectite.

Mixed-layer chlorite-expandables (R=1) have been since recently considered as discrete phases formed due to a discontinuous reaction (Meunier et al., 1988; Shau et al., 1990; Jiang & Peacor, 1994; Buatier et al., 1995; Schiffman

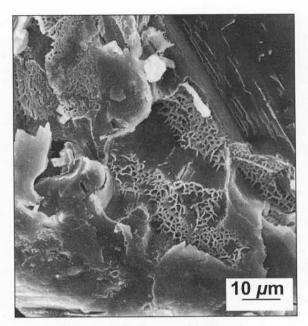


Fig. 18. Early stage of the interstratified kerolite-stevensite in amphibole-chlorite schist, sample 11a adjacent to sample 11, SEM, photo Andrzej Rochewicz.

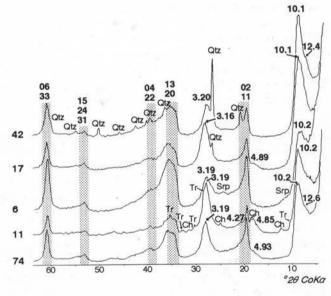


Fig. 19. Experimental X-ray tracings of samples containing kerolite-pimelite minerals (samples 6, 17, 42, 74) and interstrafied kerolite-stevensite (sample 11), non-oriented aggregates; Ch – chlorite, Qtz – quartz, Srp – serpentine, Tr – tremolite, stippled bars show *bk* bands of kerolite-pimelite.

& Staudingel 1995; Beaufort et al., 1997; Murakami et al., 1999; Roberson et al., 1999). This model seems to conform well to the origin of the interstratified chlorite-smectites from Szklary as suggested by the common occurrence of mixed-layer minerals consisting of 1:1, 2:1 and 1:2 ratios of chlorite and smectite layers, analogously to the interstratified chlorite-smectite from altered volcanoclastic rocks reported by Inoue (1987) and Inoue & Utada (1991).

The interstratified chlorite-smectite from Szklary is

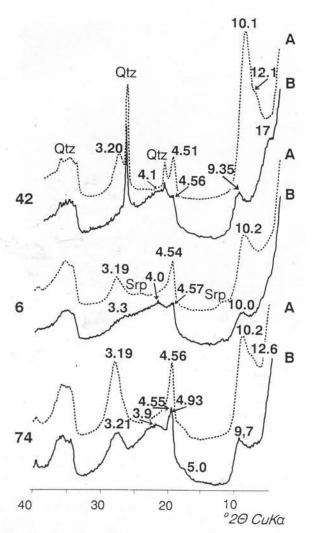


Fig. 20. Experimental X-ray tracings of kerolite-pimelite rich samples (samples 6, 42, 74), non-oriented aggregates; A – ethylene glycol treated specimen, B – air-dried specimen, Qtz – quartz, Srp – serpentine.

often accompanied by intergradient chlorite-vermiculite (intergrade; Wiewióra, 1978; Dubińska, 1982b; Wiewióra & Dubińska, 1987). The structure of the intergradient minerals is commonly regarded as a chlorite-like layer with a discontinuous brucite-like sheet or expandable layers (smectite and vermiculite) containing hydroxy islands in the interlayer space (also known as pillared structure or hydroxy interlayered vermiculite, e.g. Barnhisel & Rich 1966; Brydon & Kodama, 1966; Rich, 1968; Carstea et al., 1970; Brindley & Kao, 1980; Bain et al., 1990; Hsu, 1992; Violante et al., 1998). Intergradient minerals origin is usually ascribed to chlorite degradation (partial dissolution of a brucite-like sheet) or to the development of hydroxy islands in expandable interlayers (April, 1981; Aurousseau et al., 1983; Proust I, 1986; Buurman et al., 1988; Righi et al., 1993; Nieto et al., 1994; Carnicelli et al., 1997; Ezzim et al., 1999).

Wiewióra and Szpila (1975) characterized nickel-rich corrensite (24.12 wt. % NiO) from Szklary, with Ni²⁺-ions preferentially located in the brucite-like sheet

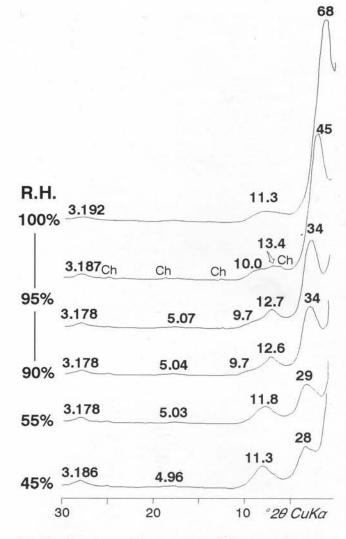


Fig. 21. Experimental X-ray tracings of Na-saturated sample 11 recorded at different relative humidities, after Wiewióra *et al.* (1982); Ch – chlorite, R.H. – relative humidity.

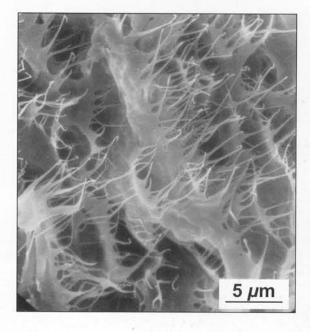


Fig. 22. Hypha-shaped aggregates of sepiolite, sample 7, SEM, photo Andrzej Rochewicz.

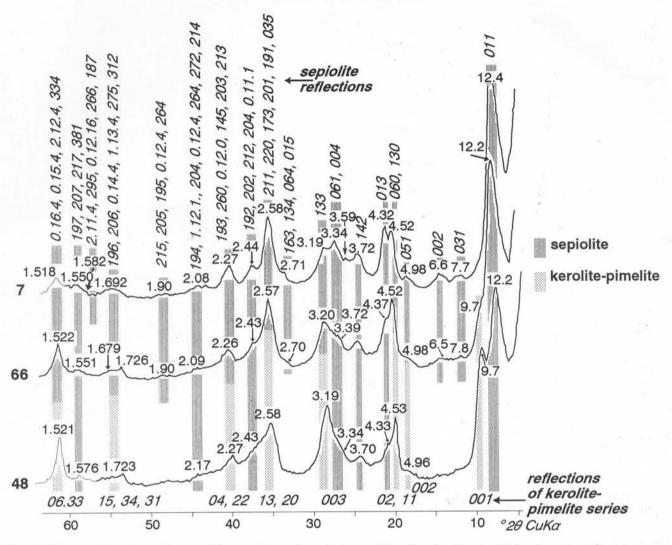


Fig. 23. X-ray tracings of samples containing sepiolite and an admixture of kerolite-pimelite, air-dried and unoriented specimens; sepiolite indices after Brindley (1959), kerolite-pimelite indices after Brindley et al. (1979).

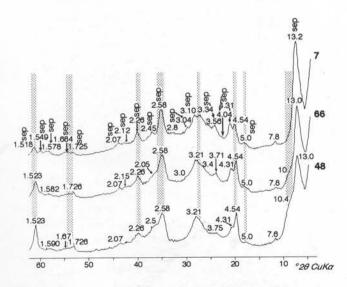


Fig. 24. X-ray tracings of samples containing sepiolite and an admixture of kerolite-pimelite, unoriented specimens treated with ethylene glycol; sep – sepiolte, stippled bands – reflections of the kerolite-pimelite series.

(Wiewióra & Anulewicz, 1976). The occurrence of corrensite and other interstratified chlorite-expandables with the asymmetric distribution of nickel suggest that continuous evolution of chlorite and vermiculite resulted in the intergrade type structure, thus both continuous and discontinuous reactions of layer silicate formation were possible in the weathering crust of Szklary.

FINAL REMARKS

The expansive set of nickeliferous layer silicates including; kerolite-pimelite series, Ni-serpentines, Ni-bearing nontronite, intergradient minerals derived form chlorite and trioctahedral mica (via vermiculitic stage) are generally known from laterites of New Caledonia type, however, the occurrence of all the above within one geological body was not reported before now. Among the inventory of minerals only kerolite-pimelite, Ni-serpentine and Ni-bearing nontronite play a significant economic

Table 4

01 1		6	9 9				12
Chemical	compositions	ot	kerol	ite-	oime	lite	series

sample no.	74	11	111	172	63	424	15	2 ⁵	35
SiO ₂	59.57	59.56	65.66	53.25	52.97	49.13	54.63	52.3	50.6
TiO ₂	0.02	0.02	0.02				0.01	0.07	0.08
Al_2O_3	0.06	1.03		0.47	0.80	1.50	0.10	0.06	0.04
Cr ₂ O ₃				tr	tr		n.d.	n.d.	n.d.
Fe ₂ O ₃	0.78	1.57	1.47	tr	tr	-	0.12	0.13	0.28
FeO	tr	0.38	0.20				× =	n.d.	n.d.
MnO	tr	0.01	0.01	- S ± ≤			2	4	1
NiO	4.82	1.08	1.21	17.99	21.97	32.61	17.97	26.1	32.7
MgO	27.8	28.56	30.19	19.8	16.07	7.40	19.11	11.15	6.58
CaO	0.85	0.57	0.64	283	(*)	0.27	0.02	0.17	0.24
K ₂ O	0.08	0.19	0.21		(4)		0.10	0.08	0.04
Na ₂ O	0.09	0.13	0.15	0.08	127	0.01	0.11	0.08	0.06
ign. loss.	6.05	6.18		5.82	8.16	9.08	7.82	8.27	8.02
total	100.12	99.48	100.00	97.41	99.97	100.00	99.99	98.41	98.64
			on	the basis of O	10(OH) ₂				
Si	3.94		4.00	3.87	3.91	3.90	3.94	4.02	4.03
Al				0.04	0.07	0.14	0.01	0.01	0.01
Fe ³⁺	0.04		0.09				0.01	0.01	0.02
Fe ²⁺			0.01						
Ni	0.26		0.06	1.05	1.30	2.08	1.04	1.62	2.09
Mg	2.74		2.74	2.14	1.77	0.88	2.05	1.28	0.78
Ca	0.06		0.04						
K	0.01		0.02						
Na	0.01		0.02	0.01			0.01		
Σ tetrahedral cat.	3.94		4.00	3.91		4.00	3.96	4.02	98.64
Σ octahedral cat.	3.04		2.90	3.19		3.00	3.09	2.92	2.89
Σ octahedral cat. corrected ⁶	3.00			3.17		2.96			
CEC meq/100g ⁷ layer charge ⁸	14.6		n.d.	29.7	32.4	21.34	n.d.	n.d.	n.d.

1. analysis corrected assuming total Al_2O_3 content is related for the admixture of chlorite of Sz69(1) composition and normalized on 100%; 2. analysis corrected for the admixture of quartz (8.9%); 4. analysis corrected for the admixture of serpentine (15.4%); 5. kerolite-pimelite minerals described by Ostrowicki (1967); 6. sum of octahedral cations corrected for CEC (cation exchange capacity); 7. CEC determined on the basis of Sr^{2+} sorption; 8. layer charge calculated on the basis of CEC; samples 6, 17 and 42 described by Wiewióra *et al.* (1982); tr – trace, n.d. – not determined

role as nickel-carriers (de Chételat, 1947, Pecora et al., 1949; Faust, 1966; Brindley & Hang, 1973; Brindley & Souza, 1975a, b; Brindley & Wan, 1975; Brindley et al., 1977; Bish & Brindley, 1978; Brindley, 1978; Durst, 1978; Maksimović & Bish, 1978; Brindley et al., 1979; Golightly & Araciba, 1979; Besset, 1980; Ducloux, 1981; Manceau & Calas, 1983; Colin et al., 1985; Manceau & Calas, 1985; Manceau et al., 1985). Most of the above listed minerals, known as classic lateritic nickel ore, can be found in the Szklary weathering crust. The list of the Szklary nickeliferous clay minerals also includes Ni-corrensite and Nirich mixed-layer serpentine-smectite unidentified in other occurrences.

The remarkable variety of phyllosilicates reflects the unusual parent rock heterogeneity and very complex origin of the Szklary massif. Layer silicate formation was concomitant to most of the geological events, starting from lizardite growth, related to the Paleozoic ocean floor metamorphism of ultramafic rocks, to the different nickel-bearing clay minerals and kaolinite related to Tertiary chemical weathering.

Layer silicates can help to identify lithologies significant for revealing geodynamic settings in which they have formed. Dioctahedral smectite occurrences are related to former mafic dykes (at present amphibolite or mafic granulite), while clintonite is always associated with metarodingites, i.e. the metamorphic equivalents of metasomatic by-products of serpentinization that can be correlated with the early serpentinization (ocean floor metamorphism) of the pristine ultramafic rocks of the Szklary massif. Intergradient minerals formed due to the alteration of chlorite and trioctahedral mica as well as massive talc occurrences are genetically related to the metasomatic zones developed at the contacts of granite-type Variscan

(?) apophyses and ultrabasic rocks.

An initial layer silicate assemblage of the contact zones plausibly produced vermiculite from the trioctahedral mica before the Tertiary chemical weathering started, as evidenced by the similar contact zone at Wiry. It may be considered an equivalent of the predecessor of the silicate nickel ores known from the weathered ultramafic rocks at Szklary (Dubińska et al., 1995; Bylina, 1996). The contact zone mineralogy and fluid inclusion data for the silica group minerals from Szklary (homogenization temperature from 70 to 120°C, Kozłowski & Sachanbiński, 1984) document equivocal hydrothermal activity of unknown age that proceeded chemical weathering. The altered vermiculite-bearing contact schists were most likely a structural and geochemical trap for nickel released from the ultramafic rocks during this hydrothermal activity (e.g. Petrenko et al., 1974; Ducloux et al., 1993) and the Tertiary weathering of the Szklary massif. This hydrothermal activity could also have resulted in the immense thickness (more than 100 m) of the original altered zone at Szklary, which is usually known as a weathering profile.

The Szklary massif is an unusual mineralogical site owing to the admirable diversity of minerals and rocks within an area of only 10 km2. Taking into account the

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Over two hundred years of ongoing mineralogical and petrological study of material from Szklary has not yet revealed all its secrets and new mineral species may still find there. The authors recently identified biopyribole developed from the anthophyllite. The biopyribole from Szklary is composed of irregularly stacked double-(amphibole) and triple-chain polysomes. This is the first report of a biopyribole occurrence in Central Europe.

The well-exposed walls of the inactive nickel ore mine at Szklary are still a valuable source of information on the origin and evolution of laterite nickel ores, and particularly information on the significance of nickel enrichment in silicates involved in forming nickel deposits of economic value. The role of metamorphic, hydrothermal, and hypergenic processes in the development of lateritic nickel deposits is far from being fully understood. The socalled weathering crust at Szklary contains a great deal of potentially fruitful material for geological and mineralogical analysis that can answer many questions and dispel many doubts.

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