Clintonite-bearing assemblages in chondrodite marbles from the contact aureole of the Třebíč Pluton, Moldanubian Zone, Bohemian Massif

Asociace obsahující clintonit v chondroditových mramorech moldanubika z kontaktní aureoly třebíčského plutonu, Český masiv

(6 figs, 4 tabs)

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Clintonite is a minor to accessory mineral in chondrodite marbles. They represent a rare type of metacarbonate rocks in the Varied Unit of the Moldanubian Zone, forming thin bodies enclosed in migmatites. Clintonite occurs exclusively in marbles from contact aureole of melanocratic ultrapotassic granites (durbachites) of the Třebíč Pluton. Chondrodite marbles consist of dominant calcite, less abundant dolomite; amounts of silicates vary from ~ 5 to 30 vol. %. The early mineral assemblage Dol+Cal+Prg ±Phl is replaced by the assemblage Chn+Cli+Cal ±Chl I ±Spl. Accessory minerals include fluorapatite, diopside, tremolite, pyrrhotite, and rare zircon and baddeleyite. Violet fluorite occurs on late fissures. Clintonite forms colourless to pale green flakes and sheaf-like aggregates, up to 2 mm in size. It has extraordinary high Si (2.739–2.986 apfu) and Si/Al ratio (0.52–0.60). The contents of Fe_{tot} (0.041–0.128 apfu), Na (0.035–0.134 apfu), Ti (0.004–0.024 apfu) and K (≤ 0.005 apfu) are low. High concentrations of F (0.437–1.022 apfu) corresponding up to 26 % of the F-component are the highest ever-recorded in clintonite. Mineral reactions producing clintonite, which is closely associated with more abundant chondrodite (X_F = 0.45–0.67), are not clear in detail, because textural relations often do not show any apparent replacement features. The simplified reactions involving diopside, pargasite and/or phlogopite: (5) Dol + Phl + Di + H_2O = Cli + Chn + Chl I + Cal + CO_2 + K_2O, (6,7) Dol + Prg (and/or Phl) = Cli + Chn + Cal + CO_2 + K_2O ± Na_2O, and (8) Dol + Phl (and/or Prg) + SiO_{2aq} + F = Cli + Chn + Spl + Cal + CO_2 + K_2O ± Na_2O seem the most probable. Retrograde chlorite II formed by the reaction: (9) Chn + Cli + CO_2 = Chl + Cal. Mineral assemblages of clintonite-bearing chondrodite marbles are product of regional-scale contact ("periplutonic") LP metamorphism at: P ~ 2–4 kbar; T ~ 620–730 °C. The reactions might be buffered internally, if pargasite and/or phlogopite were a source of F and Al; however, high modal content of chondrodite in most samples is likely related to influx of H_2O, Si and F from an external source. It is also supported by isotopic composition of calcite ($\delta^{13}C_{calcite}$ = -0.6 – -4.2 ‰ PDB; $\delta^{18}O_{calcite}$ = 12.5–15.7 ‰ SMOW). Fluorine-rich fluids very likely stabilized the assemblage chondrodite + clintonite relative to compositionally equivalent assemblages involving Mg-silicate (forsterite and/or clinohumite) + chlorite (and/or spinel) + calcite.

Key words: clintonite; chemical composition; fluorine; chondrodite marble; Moldanubian Zone; Bohemian Massif

1. Introduction

Clintonite is a rare trioctahedral brittle mica with the ideal composition $Ca(Mg_2Al)_3(SiAl_3)O_{10}(OH)_2$. It occurs chiefly in contact metamorphosed dolomite marbles, calcic and magnesian skarns (e.g. Burnham 1959, Shabynin 1973, Bucher-Nurminen 1976, Rice 1979, Bucher – Frey 1994, Sugaki *et al.* 2000, Woodford *et al.* 2001) and rarely in rodingites (Rice 1983, Dubińska 1997, Dubińska et al. 2004). Ackermand *et al.* (1986) described for the first time clintonite from regional metamorphic rock with the assemblage Cal+Ol+Cpx+Spl+Sph+Chl. More recently, Kato *et al.* (1997) described clintonite in equilibrium with primary garnet and from symplectites between garnet and clinohumite in eclogite included in marble of the UHP Su-Lu terrane, China.

Phase relations and mineral reactions of clintonite were only sporadically studied experimentally (e.g., Olesch – Seifert 1976, Zharikov – Khodorevskaya 1987). It exhibits a wide stability field in PTX diagrams but restricted to low X_{CO2}. Clintonite also requires specific chemical composition characterized by very high Al/Si ratio and low activity of Na and K (Olesch – Seifert 1976, Rice 1983, Ulmer 1983, Ackermand *et al.* 1986, Alietti *et al.* 1997, Mottana *et al.* 2002). Its crystal chemistry was studied in detail by MacKinney *et al.* (1988), but the authors did not provide any information about mineral assemblages of the studied clintonite samples. Only a low number of chemical analyses of clintonite have been published up to now and the data concerning concentrations of F and valence of Fe are rather exceptional (Rice 1979, MacKinney *et al.* 1988, Alietti *et al.* 1997, Grew *et al.* 1999). Based on the mineral assemblages and textural relations, mineral reactions producing clintonite involve calcite, spinel, forsterite and diopside (Rice 1979), less commonly also phlogopite, pargasite, chlorite and monticellite (Bucher-Nurminen 1976, Ulmer 1983).

Novák – Houzar (1996) published the first comprehensive description of clintonite closely associated with chondrodite from marbles at Tasov (Fig. 1), and two new occurrences were found later in similar marbles at Sokolí and Číhalín. All these localities of clintonite are situated in migmatites within contact aureole of the Třebíč Pluton (Houzar – Novák 2006). Here we present data concerning mineral assemblages, crystal chemistry of clintonite and associated minerals, and geological position of clintonite-bearing chondrodite marbles.

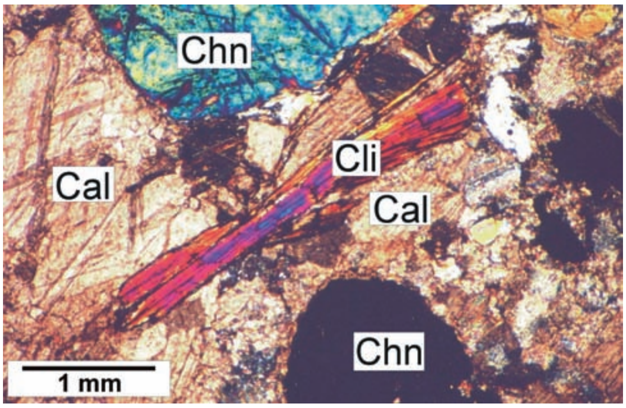

Fig. 1 Sheaf-shaped aggregates of clintonite enclosed in calcite, with isometric grains of chondrodite. Tasov near Třebíč, crossed polarizers.

2. Geologic setting

The examined chondrodite marbles occur in the Varied Unit of the Moldanubian Zone (Fig. 2). This unit is built dominantly by metapelites with the mineral assemblages Bt+Qtz+Pl+Kfs ±Sil ±Grt ±Crd, which are migmatized to a variable degree, locally with dominance of metatect over melanosome characterized by the assemblage Kfs+Qtz+Pl+Bt ±Grt ±Crd. Metapelites contain common intercalations of various rocks: calcite and dolomite marbles, locally diopside gneisses (Di+Qtz+Pl+Cal+Ttn ±Phl ±Amf), a variety of calc-silicate rocks (Di+Pl+Kfs+Ttn ±Wo ±Grs ±Cal), graphite quartzites (Qtz+Gr+Kfs+Sil+Ms ±Bt ±Pl) and amphibolites (Amp+Pl+Di+Ttn).

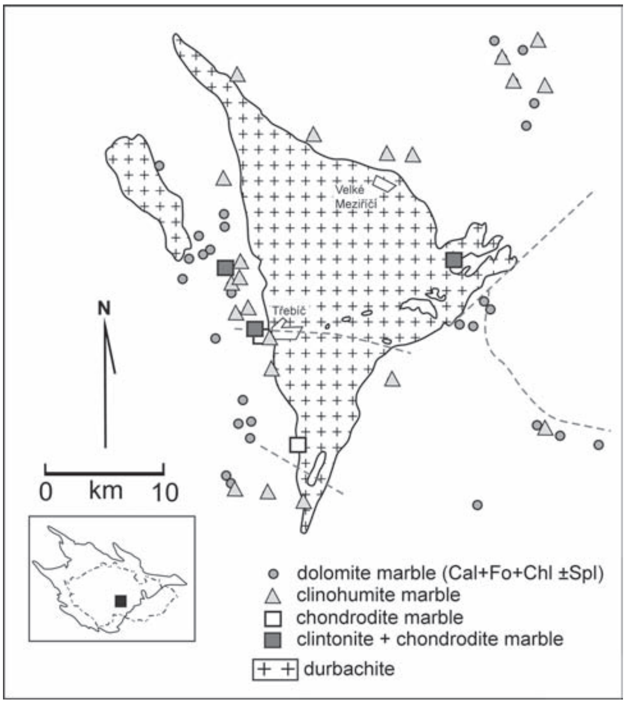

Fig. 2 Geological situation of dolomite marbles in the Třebíč Pluton area.

Several distinct types of marbles were distinguished in the eastern part of the Moldanubicum within the area of the Třebíč Pluton (Houzar 2004, Novák 1989). Abundant Si-poor dolomite marbles (15–22 wt.% MgO) with the mineral assemblage Dol+Cal+Phl+Tr+Fo+Chu+Spl+Chl, less abundant calcite marbles (< 3 wt.% MgO) with the mineral assemblage Cal+Phl+Tr+Di+Qtz+Kfs+Wo+Ttn, and rare chondrodite marbles (< 8 wt. % MgO) with the assemblage Cal+Dol+Phl+Prg+Chn+Di+Spl+Chl are documented.

Three distinct metamorphic events were recognized in the dolomite marbles, the only metacarbonate rocks studied in detail in this region. (i) The early event is represented by the assemblages: Cal+Dol+Phl+Tr ± Prg ±Di developed in almost all marbles; these minerals seem to be locally in equilibrium. They may have formed at about $T_{min.}$ = 660 °C for P = 600 MPa (Novák 1989, Houzar 2004). (ii) The dominant metamorphic event is represented by the assemblages: Cal+Dol+Fo+Chl I ±Spl ±Chu, with accessory Mg-ilmenite, geikielite and rare baddeleyite, occurring almost exclusively in dolomitic marbles. This regional-scale contact LP metamorphism (Kerrick 1991) exhibits well-developed zoning spatially related to the Třebíč Pluton (Novák – Houzar 1996). The localities of clintonite examined are located within the clinohumite zone characterized by the equilibrium mineral assemblages: Dol+Cal+Chu+Spl ± Fo ±Chl I. These assemblages are locally univariant and exceptionally invariant (Novák – Houzar 1996, Houzar 2004). The PTX conditions of this event were estimated at T = 540–620 °C for P_{fluid} 200 MPa, X_{CO2} = 0.1–0.4; Novák – Houzar 1996), or $T_{max.}$ = 660–750° for P_{fluid} = 300–400 MPa (Houzar 2004). Chlorite II was formed in a final stage of this event. (iii) Retrograde stage of metamorphism is represented by formation of abundant antigorite instead of forsterite at T < ~ 450 °C and very low X_{CO2} < 0.05 (Houzar – Novák 2001).

The Třebíč Pluton occurring in close vicinity of all clintonite localities belongs to the ultrapotassic plutonic rocks (MgO > 3 wt.%, K_2O/Na_2O > 2; Foley *et al.* 1987) of the durbachite series. The bulk composition of the Třebíč Pluton is characterized by a metaluminous signature (ASI = 0.85–0.93), high contents of K_2O (5.2–6.5 wt.%), MgO (3.3–10.4 wt.%), P_2O_5 (0.47–0.98 wt.%), Rb (330–410 ppm), Ba (1100–2470 ppm), U (6.7–26.2 ppm), Th (28.2–47.7 ppm), and Cr (270–650 ppm) (Holub 1997, Holub *et al.* 1997). The radiometric dating indicates its Lower Carboniferous age of 343±6 Ma (Pb-Pb zircon; Holub *et al.* 1997), 338±4 Ma (U-Pb zircon; Kotková *et al.* 2003).

3. Methods

Chemical composition of minerals was studied on electron microprobe Cameca SX-100 (GÚDŠ, Bratislava; operator V. Kollárová). The following analytical conditions were applied: accelerating voltage 15 kV, beam diameter 5–10 μm, beam current 20 nA and counting times

20 s. The following standards and Kα lines were used: wollastonite (Si, Ca), TiO_2 (Ti), orthoclase (K), albite (Na), Al_2O_3 (Al), MgO (Mg), hematite (Fe), rhodonite (Mn), BaF_2 (F), barite (Ba), and chromite (Cr). The concentrations of F in amphiboles, micas and chondrodite were determined using a routine electron microprobe procedure, no special methods were employed. Hence, the concentrations obtained may show quite a high error of ~ 10 rel.%. (see Ottolini *et al.* 2000). High totals of cations in chondrodite may suggest presence of intimate intergrowths of humite and/or non-stoichiometric members of humite group (Ehlers – Hoinkes 1987, Satish-Kumar – Niimi 1998). Hence, for their identification the M_{Ti}/Si (M_{Ti} = Mg+Fe^{2+}_{tot}+Mn+Ti+Ca) values given by Gaspar (1992) were used. Concentrations of F and Mg in minerals are presented as the molar ratios X_F = F/(F+OH) and X_{Mg} = Mg/(Mg+Fe^{2+}_{tot}+Mn). Except clintonite (Cli) all abbreviations are according to Kretz (1983).

4. Description of localities and textural relations of the studied samples

Číhalín

Sample CHL-1 comes from marble boulders occurring on a field ~ 1 km SW of Číhalín. Chondrodite marble forms probably thin layer(s) in cordierite-biotite gneiss with common amphibolite. Marble is white, medium grained and strongly tectonically deformed with chrysotile on fractures. It is poor in silicates (< ~5 vol. % of non-carbonate fraction). Yellow grains of chondrodite, rare colourless diopside, phlogopite partly replaced by chlorite I and pargasite occur in the marble. Scarce flakes of clintonite, up to 1 mm in diameter, locally replaced by late chlorite II, are intergrown with chondrodite (Fig. 3a). Rare accessory fluorapatite and zircon were found.

Sokolí

Layer of chondrodite marble (samples SOKP-I, SOKP-II), about 30–60 cm thick, is enclosed in metatect-rich migmatite located ~1 km SE of the village. Medium-grained, silicate-rich grey marble with graphite contains sporadically thin veins consisting of green diopside and minor quartz. Close to the contact with the migmatite, graphite-free marble is white and contains low amount of silicates. Chondrodite forms light yellow anhedral grains, 1–5 mm in diameter, and rarely up to 1 cm large, enclosed in calcite. It shows locally polysynthetic twinning. Flakes of clintonite, 0.3–1 mm in diameter, are closely associated with chondrodite, and commonly replaced by late colourless chlorite II (Fig. 3b). It locally forms complete pseudomorphs after clintonite. Rather common sheets of colourless, yellowish-brown and greenish phlogopite, up to 5 mm in diameter, are developed on foliation planes of marble. It also occurs as small inclusions in calcite, chondrodite and chlorite. Pargasite forms small colourless long prismatic crystals locally overgrown by chondrodite, or minute greenish-grey lenticular aggregates, up to 3 cm in size. Colourless diopside occurs as sporadic isolated grains and small veins. Euhedral octahedrons of violet spinel, ≤ 0.2 mm in size, microscopic accumulations of fluorapatite and scarce late violet fluorite were found in the marble.

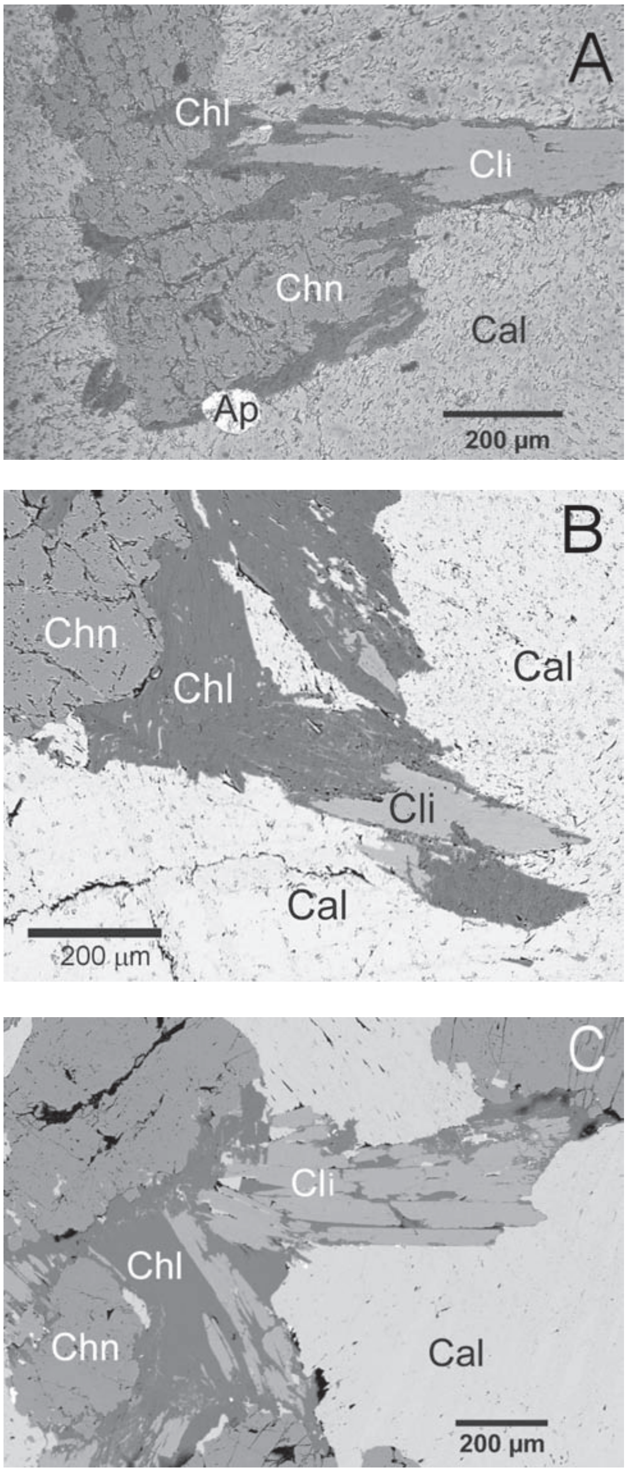

Fig. 3 BSE images of the assemblage clintonite+chondrodite+ +calcite+chlorite II: A) Číhalín, B) Sokolí, C) Tasov.

Tasov

Chondrodite marbles (samples TAS-I, TAS-II) were found in the borehole no. 179 (Uranový průzkum Company) in the depth ~ 500 m. Marbles are hosted in migmatized garnet-biotite gneisses with rare small bodies of amphibolite and serpentinite. This rock complex is located below tabular body of durbachite of the Třebíč Pluton. Chondrodite marbles most probably form a small portions or bands rich in silicates (> ~ 30 vol. % of non-carbonate fraction) in large calcite-dolomite marble body, ~ 20 m thick. Clintonite forms colourless and greenish sheaf-shaped aggregates, about 0.5–3 mm long, enclosed in or closely associated with chondrodite (Fig. 3c). It is replaced by chlorite II along contact with chondrodite, whereas clintonite enclosed solely in calcite is always fresh. Chondrodite commonly overgrows phlogopite, chlorite I and pargasite, which are locally found as tiny inclusions. It also forms lobate grains, veinlets on intergranulars connecting in minute aggregates. Accessory green spinel forms euhedral octahedral crystals, ≤ 0.4 mm in size. Chondrodite-bearing white marbles evidently replace darker marbles with common aggregates of long-prismatic greyish-green pargasite (or edenite) accompanied by less abundant phlogopite. Thin coatings of violet fluorite+calcite on marble fractures represent the latest mineralization.

The studied calcite-dominant chondrodite marbles contain low to moderate amount of dolomite (< 8 wt. % MgO). Their foliation is commonly poorly observable. The amount of non-carbonate fraction varies between < ~5 % (sample CHL-1) to ~30 % (sample TAS-1). The complete mineral assemblages of clintonite commonly comprise calcite, dolomite, phlogopite, pargasite, rarely chlorite I, diopside, and several accessory minerals (Table 1). However, only calcite, chondrodite, chlorite I, and spinel were found in direct contact with clintonite. The isotopic composition of calcite from the assemblage Cal+Chn+Cli shows, compared to average sedimentary carbonates, rather lower values: in Sokolí $\delta^{13}C$ = -0.6 to -0.5 ‰ (PDB), $\delta^{18}O$ = 17.3 to 15.7 ‰ (SMOW); in Tasov $\delta^{13}C$ = -4.2 ‰ (PDB), $\delta^{18}O$ = 12.5 ‰ (SMOW). Calcite-dolomite thermometer data (Powel *et al.* 1984) indicate that the mineral assemblages involving clintonite originated at $T_{max.}$ = 620–730 °C (Houzar 2004).

Table 1 Minerals in the individual samples of chondrodite marbles containing clintonite.

Locality sample		**Číhalín CHL-1**	**Sokolí SOKP-1**	**Sokolí SOKP-2**	**Tasov TAS-1**	**Tasov TAS-2**
Dolomite	**Dol**	x	x	X	x	X
Calcite	**Cal**	X	X	X	X	X
Phlogopite	**Phl**	t	X	X	t	x
Pargasite	**Prg**	t	x	x	x	X
Edenite	**Ed**	-	-	-	-	x
Chlorite I	**Chl**	X	x	-	-	-
Clintonite	**Cli**	t	x	x	x	t
Chondrodite	**Chn**	X	X	X	X	X
Tremolite	**Tr**	-	t	-	-	t
Diopside	**Di**	x	t	x	-	-
Spinel	**Spl**	-	t	x	X	x
Graphite	**Gr**	-	-	t	-	-
Fluorapatite	**Ap**	t	x	t	t	t
Fluorite	**Fl**	-	t	-	t	t
Zircon	**Zrn**	t	-	-	-	-
Baddeleyite	**Bad**	-	-	-	t	-
Sphalerite	**Sp**	-	-	-	t	-
Arsenopyrite	**Apy**	-	-	-	-	t
Pyrhotite	**Po**	-	-	-	t	x
Scheelite	**Sch**	-	-	t	-	-
Chlorite II	**Chl II**	X	X	X	x	x
Serpentine	**Srp**	X	-	-	-	-

X = major constituent, **x** = minor constituent, **t** = trace
Abbreviations from Kretz (1983) except Cli for clintonite and Sch for scheelite

5. Chemistry of minerals from clintonite-bearing chondrodite marbles

Clintonite

Chemical composition of clintonite with X_{Mg} = 0.97–0.99 exhibits very high contents of Si (2.739–2.986 apfu), which is typical for clintonite from marbles, Al^{IV} is rather low (5.014–5.261 apfu) as a result of the substitution $^{IV}Al_{-1}{}^{VI}Mg_{-2}{}^{IV}Si^{VI}Al^{VI}\square$ (Alietti *at al.* 1997, Mottana *et al.* 2002). The ratio Si/Al^{IV} (0.52 to 0.60) is much higher than the ideal value (Si/Al^{IV} = 0.33). The contents of Fe^{2+}_{tot} (0.041–0.128 apfu) and Na (0.035–0.134 apfu) are low and variable, the highest values were found in Tasov (Table 2). Concentration of Ti is very low (0.004–0.024 apfu) as well as K (≤0.005 apfu) and Cr (0.001–0.012 apfu), the contents of Mn and Ba were below detection limit. Clintonite exhibits high contents of F (0.437–1.022 apfu, X_F = 0.11–0.26), which represent the highest known values ever found (Fig. 4). High contents of F and high Si/Al ratios are similar to those in clintonite from fluorine-rich marbles at the Orange County localities, New York, USA (MacKinney *et al.* 1988).

Chondrodite

Chondrodite exhibits high X_{Mg} = 0.95–0.99, Fe varies from 0.077 apfu to 0.224 apfu; the highest contents of Fe are from chondrodite at Tasov. It is poor in Ti (0.012–0.043 apfu), Mn (≤ 0.008 apfu) and Ca (≤ 0.004 apfu). Variable but moderate amounts of F (X_F = 0.45–0.67) were found. Histogram representing the range of M_{Ti}/Si values shows bimodal distribution (Fig. 5). This suggests

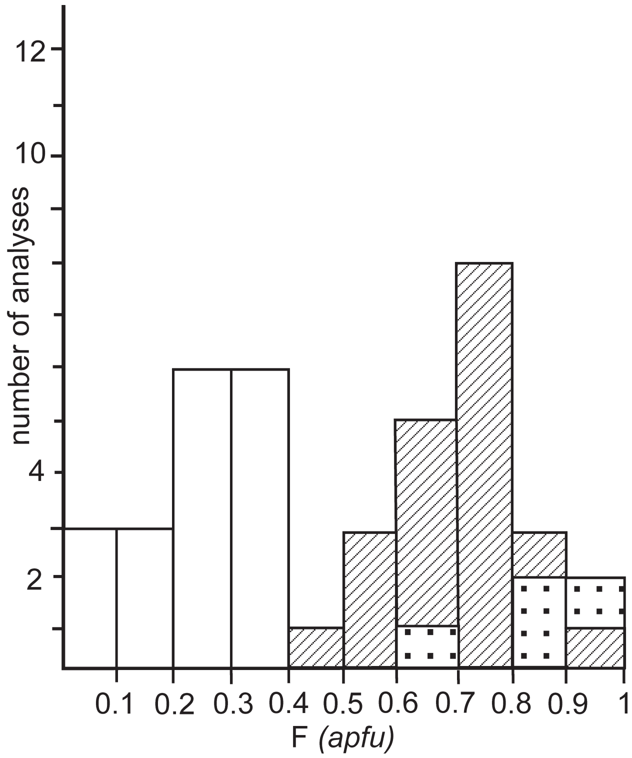

Fig. 4 Diagram showing the fluorine concentrations in clintonites: shaded – studied localities; dotted – clintonite from marbles in the Orange County, USA; white – other clintonites (data source: Rice 1979, MacKinney *et al.* 1988, Allieti *et al.* 1997, Grew *et al.* 1999, this study).

Table 2 Representative microprobe analyses of clintonite.

	CHL-I	CHL-I	CHL-I	SOKP-I	SOKP-I	SOKP-I	SOKP-II	SOKP-II	TAS-I	TAS-I	TAS-II	TAS-II
	Cli	**Cli**	**Cli**	**Cli**	**Cli**	**Cli**	**Cli**	**Cli**	**Cli**	**Cli**	**Cli**	**Cli**
SiO_2	19.57	19.68	19.99	20.19	20.00	20.24	20.44	19.91	20.04	20.54	21.00	20.71
TiO_2	0.17	0.19	0.14	0.06	0.04	0.07	0.04	0.05	0.06	0.07	0.09	0.09
Al_2O_3	39.51	39.32	38.69	38.36	39.16	38.37	38.55	37.42	37.77	38.15	37.02	36.89
Cr_2O_3	0.03	0.06	0.02	0.04	0.03	0.03	0.06	0.03	0.03	0.05	0.01	b.d.
FeO	0.72	0.80	0.70	0.48	0.57	0.35	0.49	0.41	1.01	0.91	1.07	1.08
MgO	21.81	21.94	22.40	22.82	22.34	22.84	22.34	22.49	22.29	22.57	22.44	22.45
CaO	13.23	13.12	13.31	13.09	13.29	13.29	13.57	13.23	12.81	12.9	12.83	12.81
Na_2O	0.18	0.31	0.18	0.20	0.13	0.21	0.13	0.13	0.28	0.35	0.33	0.34
K_2O	0.02	0.02	0.02	0.02	0.02	0.01	b.d.	0.01	0.02	0.03	0.03	0.02
H_2O *	3.51	3.52	3.46	3.47	3.82	3.67	3.41	3.51	3.48	3.43	3.38	3.15
F	1.60	1.59	1.73	1.71	0.99	1.29	1.86	1.46	1.57	1.81	1.84	2.28
O=F	-0.67	-0.67	-0.73	-0.72	-0.42	-0.54	-0.78	-0.61	-0.66	-0.76	-0.77	-0.96
TOTAL	99.68	99.89	99.91	99.72	99.98	99.83	100.11	98.04	98.70	100.05	99.27	98.86
Si^{4+}	2.747	2.758	2.800	2.829	2.794	2.832	2.854	2.839	2.843	2.873	2.961	2.936
Al^{IV}	5.253	5.242	5.200	5.171	5.206	5.168	5.146	5.161	5.157	5.127	5.039	5.064
Al^{VI}	1.282	1.253	1.188	1.165	1.241	1.159	1.199	1.129	1.159	1.162	1.113	1.099
Ti^{4+}	0.018	0.020	0.015	0.006	0.004	0.007	0.004	0.005	0.006	0.007	0.010	0.010
Cr^{3+}	0.003	0.007	0.002	0.004	0.003	0.003	0.007	0.003	0.003	0.006	0.001	–
Fe^{2+}_{tot}	0.085	0.094	0.082	0.056	0.067	0.041	0.057	0.049	0.120	0.106	0.126	0.128
Mg^{2+}	4.563	4.584	4.678	4.768	4.652	4.764	4.651	4.782	4.715	4.707	4.717	4.744
Ca^{2+}	1.989	1.970	1.998	1.965	1.989	1.992	2.030	2.022	1.947	1.933	1.938	1.945
Na^{+}	0.049	0.084	0.049	0.054	0.035	0.057	0.035	0.036	0.077	0.095	0.090	0.093
K^{+}	0.004	0.004	0.004	0.004	0.004	0.002	–	–	0.004	0.005	0.005	0.004
H^{+}	3.290	3.295	3.233	3.242	3.563	3.429	3.179	3.341	3.296	3.199	3.179	2.978
F^{-}	0.710	0.705	0.767	0.758	0.437	0.571	0.821	0.659	0.704	0.801	0.821	1.022
O^{2-}	23.29	23.295	23.233	23.242	23.563	23.429	23.179	23.341	23.296	23.199	23.179	22.978
CATSUM	15.993	16.015	16.016	16.023	15.996	16.025	15.983	16.028	16.031	16.022	16.001	16.022
AN SUM	24	24	24	24	24	24	24	24	24	24	24	24
X_F	0.18	0.18	0.19	0.19	0.11	0.14	0.21	0.17	0.18	0.20	0.21	0.26

the possibility of presence of submicroscopic intergrowth of chondrodite with humite (cf. Rice 1980, Satish-Kumar – Niimi 1998). Representative chemical data of chondrodite are given in Table 3.

Other minerals

Phlogopite has high X_{Mg} = 0.96–0.99 and high Al (up to 2.726 apfu), low content of Ti (≤ 0.033 apfu), Fe (≤ 0.229 apfu), Mn (≤ 0.005 apfu) and Na (= 0.294 apfu), concentration of Ba is below detection limit (Table 4). Elevated contents of F (X_F = 0.20–0.39) are typical. *Pargasite* (to rare edenite) shows X_{Mg} = 0.92–0.94, variable contents of Al (1.927–2.732 apfu), Ti (0.029–0.098 apfu) and Fe (0.159–0.354 apfu), Mn is always close to the detection limit. Pargasite from Číhalín and Sokolí is relatively rich in K (≤ 0.321 apfu), locally with elevated content of F (X_F = 0.39–0.61) (Table 4). *Diopside* is close to the end-member composition with X_{Mg} = 0.98–1.0. *Spinel* exhibits moderate X_{Mg} = 0.78–0.90, slightly elevated contents of Fe (≤ 0.178 apfu) and Zn (≤ 0.043 apfu), with Mn and Cr close to the detection limits (Table 4). This Fe-rich spinel from locality Tasov has similar composition as early core of euhedral spinel from

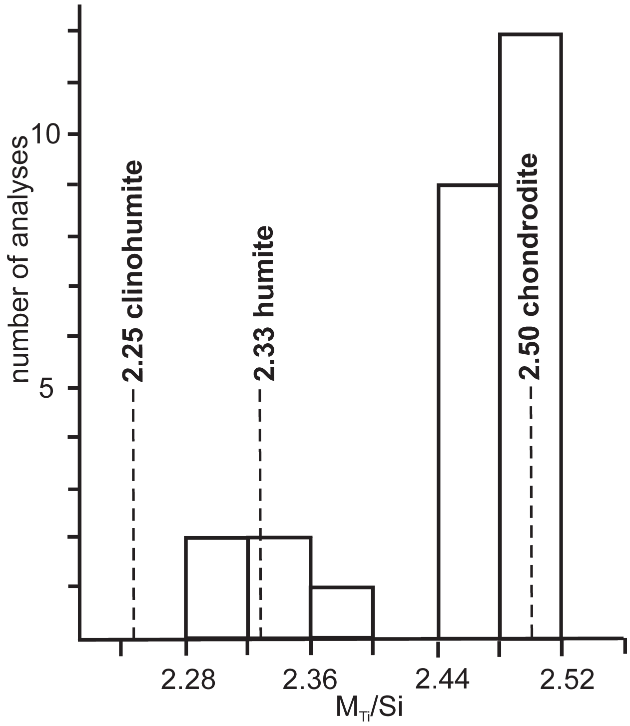

Fig. 5 Diagram showing M_{Ti}/Si ratio in studied chondrodite.

Table 3 Representative microprobe analyses of chondrodite and chlorite II.

	CHL-I Chn*	SOKP-I Chn	SOKP-II Chn	TAS-I Chn	TAS-II Chn	CHL-I Chl II	SOKP-II Chl II	TAS-I Chl II
SiO_2	37.28	34.87	35.25	35.32	34.89	32.16	31.17	30.82
TiO_2	0.59	0.30	0.38	0.27	0.31	0.07	0.02	0.05
Al_2O_3	b.d.	b.d.	b.d.	b.d.	b.d.	19.66	21.51	20.87
FeO	3.51	2.22	2.26	2.75	4.65	1.55	0.98	1.70
MnO	0.07	0.04	0.04	0.06	0.16	b.d.	b.d.	b.d.
MgO	55.21	56.98	57.17	56.53	54.99	34.09	33.68	33.31
CaO	0.05	0.02	b.d.	0.07	0.04	0.18	0.03	0.03
H_2O *	2.79	1.92	2.11	1.72	1.97	12.71	12.66	12.27
F	5.42	6.98	6.66	7.46	6.83	0.56	0.68	1.17
O=F	-2.28	-2.94	-2.80	-3.14	-2.88	-0.24	-0.29	-0.49
TOTAL	102.63	100.39	101.08	101.04	100.96	100.95	100.52	99.73
Si^{4+}	2.087	1.998	2.005	2.015	2.009	2.969	2.880	2.882
Ti^{4+}	0.025	0.013	0.016	0.012	0.013	0.005	0.001	0.004
Al^{3+}	–	–	–	–	–	2.139	2.342	2.300
Fe^{2+}_{tot}	0.164	0.106	0.107	0.131	0.224	0.120	0.076	0.133
Mn^{2+}	0.003	0.002	0.002	0.003	0.008	–	–	–
Mg^{2+}	4.607	4.868	4.848	4.808	4.721	4.692	4.639	4.643
Ca^{2+}	0.003	0.001	–	0.004	0.002	0.018	0.003	0.003
H^+	1.041	0.735	0.802	0.654	0.756	7.826	7.800	7.652
F^-	0.959	1.265	0.198	1.346	1.244	0.164	0.199	0.346
O^{2-}	9.041	8.735	8.802	8.654	8.756	17.826	17.800	17.652
CATSUM	6.889	6.989	6.979	6.973	6.977	9.964	9.947	9.967
AN SUM	10	10	10	10	10	18	18	18
X_F	0.48	0.63	0.60	0.67	0.61			

* including probable humite

Table 4 Representative microprobe analyses of pargasite, phlogopite, diopside and spinel.

	CHL-I Prg	SOKP-I Prg	TAS-II Prg	CHL-I Phl	SOKP-I Phl	SOKP-II Phl	TAS-II Phl	CHL-I Di	SOKP-I Spl	AS-I cor Spl	TAS-I rim Spl
SiO_2	43.42	42.48	43.15	41.12	41.31	40.79	42.06	55.31	b.d.	b.d.	b.d.
TiO_2	0.92	0.27	0.81	0.28	0.14	0.19	0.28	0.05	b.d.	b.d.	b.d.
Al_2O_3	15.05	15.06	14.23	15.85	15.37	15.68	14.87	0.17	69.64	67.46	67.45
Cr_2O_3	b.d.	b.d.	b.d.	b.d.	0.04	0.03	b.d.	b.d.	0.19	b.d.	b.d.
Fe_2O_3									1.07	2.13	1.45
FeO	1.37	2.43	2.97	1.02	0.77	0.67	1.97	0.85	3.62	7.42	8.59
MnO	0.03	b.d.	b.d.	0.03	0.04	0.02	b.d.	0.02	0.04	b.d.	b.d.
ZnO	b.d.	b.d.	b.d.	b.d.	b.d.	b.d.	b.d.	b.d.	1.44	1.66	2.32
MgO	18.81	20.28	18.70	25.74	27.35	27.04	25.92	18.00	25.11	22.22	21.06
CaO	13.93	13.66	13.48	b.d.	b.d.	b.d.	b.d.	25.98	b.d.	b.d.	b.d.
Na_2O	2.21	1.62	3.18	0.84	0.22	0.21	1.09	0.05	b.d.	b.d.	b.d.
K_2O	1.50	1.23	0.96	10.06	10.43	10.49	9.47	b.d.	b.d.	b.d.	b.d.
H_2O *	1.22	1.10	1.47	2.66	3.18	3.34	3.29				
F	1.88	2.12	1.25	3.43	2.40	2.01	2.16				
O=F	-0.79	-0.89	-0.53	-1.44	-1.01	-0.85	-0.91				
TOTAL	99.55	99.35	99.80	99.58	100.24	99.62	100.2	100.43	101.12	100.89	100.88
Si^{4+}	6.162	6.059	6.158	5.756	5.739	5.701	5.849	1.994	–	–	–
Ti^{4+}	0.098	0.029	0.087	0.029	0.015	0.020	0.029	0.001	–	–	–
Al^{3+}	2.517	2.532	2.393	2.615	2.516	2.583	2.437	0.007	1.976	1.960	1.973
Cr^{3+}	–	–	–	–	0.004	0.003	–	–	0.004	–	–
Fe^{3+}									0.019	0.040	0.027
Fe^{2+}	0.163	0.290	0.354	0.119	0.089	0.078	0.229	0.026	0.073	0.153	0.178
Mn^{2+}	0.004	–	–	0.004	0.005	0.002	0	0.001	0.001	–	–
Zn^{2+}	–	–	–	–	–	–	–	–	0.026	0.030	0.043
Mg^{2+}	3.980	4.312	3.978	5.371	5.664	5.634	5.373	0.967	0.901	0.817	0.779
Ca^{2+}	2.018	2.088	2.061	–	–	–	–	1.003	–	–	–
Na^{+}	0.608	0.448	0.880	0.228	0.059	0.057	0.294	0.003	–	–	–
K^{+}	0.272	0.224	0.175	1.797	1.848	1.870	1.680	–	–	–	–
H^{+}	1.156	1.044	1.397	2.481	2.946	3.111	3.050	–	–	–	–
F^{-}	0.844	0.956	0.564	1.519	1.054	0.889	0.950	–	–	–	–
O^{2-}	23.156	23.044	23.397	22.481	22.946	23.111	23.050	–	–	–	–
CATSUM	15.921	15.982	16.086	15.919	15.94	15.949	15.891	4	3	3	3
AN SUM	24	24	24	24	24	24	24	6	4	4	4

dolomite marbles with the assemblage Fo+Chu+Spl in Horní Libochová (Novák 1988). Both *chlorite I* and *chlorite II* are colourless. Only chlorite II was analysed having high X_{Mg} = 0.97–0.99 with Fe (≤ 0.120 apfu) and Al contents 2.004–2.342 apfu. Chlorite II locally contains small amounts of F ≤ 0.346 apfu (Table 3).

6. Discussion

6.1. Textural relations and mineral reactions

Textural relations in clintonite-bearing chondrodite marbles enable to recognize three stages, which are characterized by the mineral assemblages (calcite+dolomite are always present): (i) pargasite+phlogopite+diopside(?); (ii) chondrodite+clintonite+chlorite I or spinel; (iii) chlorite II. Spinel as well as chlorite I, both formed by replacement of phlogopite (or pargasite) are locally absent. Position of diopside is not clear, because it is always enclosed in calcite and its contact with any other silicate mineral was not found. This sequence of mineral assemblages is very similar to that known from dolomite marbles in this region, where the following mineral assemblages were recognized in the individual stages (Cal+Dol always present): (i) tremolite+phlogopite+pargasite; (ii) forsterite+clinohumite+spinel+chlorite I; (iii) chlorite II (cf. Novák 1988, 1989, Novák – Houzar 1996, Houzar 2004).

Mineral reactions producing clintonite in marbles involve calcite, clinopyroxene (diopside), forsterite and monticellite as reactants, spinel is considered as a source of Al (Bucher-Nurminen 1975, Rice 1979, Ulmer 1983). The following reactions were given:

(1) Fo + Cpx + Spl + Cal + H_2O = Cli + CO_2

(2) Cpx + Spl + Cal + H_2O = Cli + Fo + CO_2

The reaction (2) strongly depends on X_{CO2}, particularly for T > 550 °C (Rice 1979). Bucher-Nurminen (1975) discussed a possibility of clintonite production by dealkalization of phlogopite and pargasite according to the reactions:

(3) Cal + Phl + Prg = Cli + Di + CO_2 + H_2O + K_2O + Na_2O

(4) Cal + Phl + Spl + H_2O = Cli + Fo + CO_2 + K_2O

However, dolomite as a typical mineral in these assemblages and typical reactant in prograde carbonate+silicate reactions (see e.g. Skippen 1974) is not involved in the reactions (1) to (4). Moreover, participation of minerals with elevated concentrations of F has not been discussed as well. Consequently, the reactions given above cover only a small part of natural mineral assemblages found in marbles.

The examined localities differ in their mineral assemblages and textural relations from the above examples; hence, new reactions (detailed stoichiometry is not given) have been suggested. At the Číhalín locality, textural relations indicate the reaction:

(5) Dol + Phl + Di + H_2O = Cli + Chn + Chl I + Cal + CO_2 + K_2O

At Sokolí and Tasov, the presence of pargasite and phlogopite and their textural relations suggest the reactions:

(6) Dol + Prg + H_2O = Cli + Chn + Cal + CO_2 + Na_2O ± K_2O

(7) Dol + Phl + H_2O = Cli + Chn + Cal + CO_2 + K_2O

(8) Dol + Phl (and/or Prg) + SiO_{2aq} + F = Cli + Chn + Spl + Cal + CO_2 + K_2O ± Na_2O

The abundance of chondrodite (up to ~ 25 vol. %), high chondrodite/clintonite ratios and isotopic composition of calcite support introduction of H_2O, Si and very likely also F into the system, whereas Na and K were removed. Based on detailed study of reaction textures, similar dealkalization reactions producing chlorite and/or spinel instead of clintonite, and forsterite instead of chondrodite were described for amphibole and phlogopite (Kretz 1980, Bucher-Nurminen 1982, Novák 1989). However, they were not tested experimentally and the breakdown of pargasite and phlogopite is still unclear; perhaps a high acidity of fluids may have operated (cf. Bucher-Nurminen 1982).

Retrograde chlorite II, occurring exclusively at the contact between chondrodite and clintonite, formed by the reaction:

(9) Cli + Chn + CO_2 + H_2O = Chl + Cal

The reactions (7) and (9) are very similar to those described by Novák (1989) and Novák – Houzar (1996) for dolomite marbles in the Strážek Moldanubicum, characterized by the presence of forsterite and/or clinohumite instead of chondrodite and spinel and/or chlorite I instead of clintonite.

6.2. Role of fluorine in mineral reactions

The mineral reactions producing clintonite in marbles were only exceptionally studied experimentally (Rice 1979). The assemblage chondrodite+clintonite is rare in nature and it has not been studied to date. Moreover, the reactions observed in the studied region are quite different from those examined by Rice (1979). Consequently, we can estimate PT(X) conditions of formation of the assemblage chondrodite+clintonite only from analogy with associated dolomite marbles with the assemblage Fo+Chu+Spl+Chl. Nevertheless, the estimated T ~620–730 °C for P ~ 2–4 kbar for these marbles are in agreement with the stability field of clintonite and chondrodite (Olesch – Seifert 1976, Rice 1979, 1980).

The mineral assemblages and reactions described above may suggest two distinct ways of clintonite formation controlled chiefly by activity of F. If the newly formed mineral assemblage shows low amount of chondrodite and clintonite, and low chondrodite/clintonite ratio, replaced phlogopite and/or pargasite possibly provided enough F for formation of the both F-bearing minerals, e.g., according to the reaction (5) and perhaps also reactions (6) and (7); influx of F into the system is not necessary. Novák (1988) found such a relation in a newly formed assemblage Fo+Chu+Spl+Cal replacing phlogopite and dolomite at locality Studnice in the Strážek Moldanubicum. Nevertheless, the most widespread textural relations, abundance of chondrodite (locally up to 25 vol. %), high chondrodite/clintonite ratio as well as the isotopic compositions of calcite from some samples at Sokolí and Tasov strongly suggest influx of F in external fluids, where the reaction (8) and perhaps also reactions (6) and (7) operated. Consequently, F-rich clintonite (X_F = 0.11–0.26) associated with chondrodite crystallized from external fluids introducing F, Si and H_2O but perhaps at relatively high X_{CO2} < 0.6. The modal composition and textural relations of the assemblage chondrodite + clintonite suggest dominance of the crystallization in an open system with influx of F at most localities, relative to crystallization in a closed system. The latter case may correspond only to marble domains with a low chondrodite content.

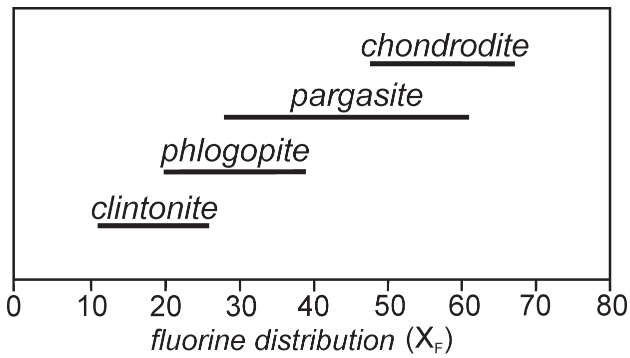

Fig. 6 Distribution of F in the minerals from the studied chondrodite marbles.

Partitioning of F into the individual minerals: X_F = Cli $_{(0.11–0.26)}$ < Phl $_{(0.20–0.39)}$ < Prg $_{(0.28–0.61)}$ < Chn $_{(0.45–0.67)}$ (Fig. 6) is similar to that found in marbles from the Franklin Marble, Orange County, USA (Kearns *et al.* 1980). Disregarding the fact that the examined clintonite is F-poor relative to the other associated minerals, it is characterized by the highest concentrations of F ever recorded in this mineral (X_F = 0.11–0.26; 1.13–2.28 wt. % F). Similar assemblages and high F contents in clintonite were found in marbles at localities Amity, Edenville and Warwick (Orange County, New York, USA; (X_F = 0.17–0.23; 1.45–2.05 wt. % F; MacKinney *et al.* 1988). This is in contradiction with a quite low amount of F in associated chondrodite (X_F = 0.45–0.67) relative to chondrodite from other localities and experimental study (Rice 1980, 1979).

7. Conclusions

Formation of clintonite is very likely related to HT/LP metamorphism in the aureole of the Třebíč Pluton. Bucher – Frey (1994) considered clintonite a typomorphic mineral in the mineral assemblages Cal+Fo+Chu+Spl+Cli and Cal+Fo+Di+Spl+Cli located in contact aureoles of granite plutons. However, why clintonite occurs only in the chondrodite marbles but it is absent in associated dolomite marbles with the assemblage Cal+Fo+Chu+Spl+Chl, very similar to those described by Bucher – Frey (1994), is unclear. Origin of clintonite seems to be related to elevated activity of F indicated also by abundant chondrodite. Its composition with X_F = 0.45–0.67 suggests elevated activity of F but also variable and high X_{CO2} < 0.6 (see Rice 1980) relative to rather low X_{CO2} < 0.4 derived from clinohumite composition and mineral relations in associated dolomite marbles with clinohumite (Novák – Houzar 1996). Activity of F seems to be the main factor controlling formation of clintonite besides general factors such as low Si/Al ratio and a low activity of K and Na.

All localities of chondrodite marbles occur within clinohumite zone developed around the Třebíč Pluton. They are enclosed in migmatites and the distance of the individual localities from the nearest outcrop of durbachite rocks at the present surface varies from ~ 0.2 to ~ 3 km. Along with the examined Moravian localities, clintonite was recently discovered by the authors at Kamenné doly quarry near Písek, southern Bohemia. This chondrodite marble forms enclave, ~ 4 m in diameter, enclosed in durbachite. It exhibits the assemblage Cal+Cli+Chn+Prg+Spl+Chl II, very similar to those described in chondrodite marbles around the Třebíč Pluton. The presence of marble enclave in durbachite supports contact-metamorphic origin of clintonite in the inner part of aureole of the Třebíč Pluton, disregarding large distance of some clintonite localities from the nearest durbachite outcrop. However, as the Třebíč Pluton forms a tabular body (Rejl – Sedlák 1987), the distance of chondrodite marbles from the nearest durbachite outlier, as estimated in geological maps, may be incorrect. The Tasov borehole documented tabular durbachite bodies overlying chondrodite marbles and indicates that similar overlying bodies of durbachite may have been removed by erosion at the other studied localities.

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Asociace obsahující clintonit v chondroditových mramorech moldanubika z kontaktní aureoly třebíčského plutonu, Český masiv

Clintonit, vzácná trioktaedrická slída s ideálním chemickým vzorcem Ca $(Mg_2Al)_3(SiAl_3)O_{10}(OH)_2$, byla zjištěna v chondroditových mramorech, uložených v migmatitech v blízkosti třebíčského plutonu (lokality Čihalín, Sokolí a Tasov). Clintonit v texturně rovnovážné asociaci s chondroditem, vzácněji se spinelem nebo chloritem I, náleží k mladší metamorfní asociaci těchto mramorů, která vznikla na úkor starší asociace Dol+Phl+Prg +Di(?) + Spl(?). Z akcesorických minerálů mramorů lze dále zmínit fluorapatit, pyrhotin, fluorit, zirkon a baddeleyit. Retrográdní stadium metamorfózy je reprezentováno vznikem hojného chloritu II na úkor clintonitu na přímém kontaktu s chondroditem. Studovaný clintonit se vyznačuje vysokým poměrem Si/Al (0.52–0.60) a zejména vysokým obsahem F (X_F = 0.11–0.26), čímž odpovídá fluorem nejbohatšímu dosud zjištěnému clintonitu. Asociující chondrodit (patrně je vzácně přítomen i submikroskopický humit) je naopak fluorem relativně chudý (X_F = 0.45–0.67). Clintonit s chondroditem mohl vznikat zčásti v uzavřeném systému reakcemi, kdy zdrojem F byl flogopit a pargasit:

(5) Dol + Phl + Di + H_2O = Cli + Chn + Chl I + Cal + CO_2 (Čihalín)

(6) Dol + Prg + H_2O = Cli + Chn + Cal + CO_2 + Na_2O ± K_2O (Sokolí, Tasov)

(7) Dol + Phl + H_2O = Cli + Chn + Cal + CO_2 + K_2O (Sokolí, Tasov)

Reálnější, a složením stabilních izotopů C a O v kalcitu chondroditových mramorů podpořený, je však vznik asociace clintonit+chondrodit za přínosu F, H_2O a SiO_2 z externího zdroje (zejména lokality Sokolí a Tasov) reakcí:

(8) Dol + Phl (and/or Prg) + SiO_{2aq} + F = Cli + Chn + Spl + Cal + CO_2 + K_2O ± Na_2O

Relativně vysoký podíl F ve fluidech patrně podporoval vznik clintonitu namísto spinelu a chloritu. Ve srovnání s dříve popsanými výskyty clintonitu v mramorech kontaktních aureol granitoidních plutonů, nevznikl studovaný clintonit na přímém kontaktu magmatitů, ale v podmínkách periplutonické LP metamorfózy v aureole třebíčského plutonu, které lze přibližně odhadovat při předpokládaném P = 200–400 MPa na T = 620–730 °C a X_{CO2} < 0.6.