TITANITE-ILMENITE-MAGNETITE PHASE RELATIONS IN AMPHIBOLITES OF THE CHÝNOV AREA (BOHEMIAN MASSIF, CZECH REPUBLIC)

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ABSTRACT

An assemblage of Al-bearing titanite, ilmenite and magnetite is present in polymetamorphic metabasite environment of the Varied group of the Moldanubian Zone. Titanite is characterized by coupled substitution (Al, Fe^{3+})+ (F, OH) ⇔ Ti + O, with a slight excess of (Al, Fe^{3+})-F component. Complex reaction rims of titanite around ilmenite occur in some cases. Ilmenites have $FeTiO_3$ amounts usually above 96 mol.%, with some enrichment by $MnTiO_3$ component (max. 8 mol.%). Magnetite has a negligible ulvöspinel component (max. 1 mol.%).

KEYWORDS: Bohemian Massif, Moldanubian Zone, amphibolite, titanite, ilmenite, magnetite

INTRODUCTION

Titanite $CaTiOSiO_4$ together with ilmenite $FeTiO_3$ and magnetite Fe_3O_4 are common accessory minerals in a wide range of igneous and metamorphic rocks. These mineral assemblages provide valuable information about variations in oxygen fugacity (fO_2) and water fugacity (fH_2O) during the evolution of geological environment (e.g., Wones, 1989; Xirouchakis et al., 2001a,b; Troitzsch and Ellis, 2002; Tropper et al., 2002; Harlov et al., 2006). Titanite, with its considerable variation in crystal chemistry, is a highly sensitive indicator of both parameters. The octahedral sites of titanite are normally occupied by Ti^{4+} (Ribbe, 1980), but Al^{3+} and Fe^{3+} may by incorporated by means of substitution $Ti^{4+} + O^{2-} \Leftrightarrow M^{3+} + (OH, F)^-$, which has been found in many natural titanites (e.g., Franz and Spear, 1985; Enami et al., 1993; Markl and Piazolo, 1999; Troitzsch and Ellis, 2002; Tropper et al., 2002; Harlov et al., 2006).

In the present study, the occurrence of (Al, Fe^{3+})-(F, OH)-bearing titanites in assemblages with ilmenites and magnetites are described from a poly-metamorhic metabasite environment of the Chýnov area. The aim of this study is to describe and discuss mineral assemblages of titanite with magnetite and ilmenite, their textural relationships and the fO_2-fH_2O conditions governing metamorphic reactions.

GEOLOGICAL SETTING

The Moldanubian Zone forms the central part of the Bohemian Massif. In the recent literature (Franke, 1989, 2000; Finger et al., 2007) two major tectonic subunits are commonly distinguished within this zone: the Gföhl Unit and the Drosendorf Unit. The Gföhl Unit, which posseses a record of Variscan high-pressure metamorphism, includes mainly felsic granulites and leucocratic migmatitic orthogneisses (often summarized as Gföhl gneisses), as well as minor amounts of amphibolites, ultrabasic rocks and paragneisses. The Drosendorf Unit sensu Franke (2000) comprises mainly high-grade metasediments with intercalations of amphibolites and orthogneisses. An opposed to the Gföhl Unit, the metamorphic pressures were in general low to medium in these rocks. According to the traditional lithostratigraphic concept (Kodym, 1966; Jenček and Vajner, 1968; Zoubek, 1988), the Drosendorf Unit can be further subdivided into the Monotonous and Varied groups. The Monotonous group is represented by paragneisses derived from psammitic-pelitic sediments (Franke, 2000; René, 2006). The Varied group includes occurrences of various metasedimentary series, which contain significant intercalations of carbonate rocks, quartzites, amphibolites and orthogneisses (Fig. 1). Amphibolites are a very significant part of Varied group rocks sequences. They probable include metabasites of different age and origin. However, Early Paleozoic origin (509±27 Ma) of amphibolites from the Chýnov area was presented by Janoušek et al. (1997).

The Chýnov area comprises an association of amphibolites with dolomitic marbles in the central part of the Varied group, northeast of Tábor (Fig. 2). The relatively abundant intercalations and irregular lenses of amphibolites, together with marbles, occur in the complex of two-mica gneisses (Vrána, 1992;

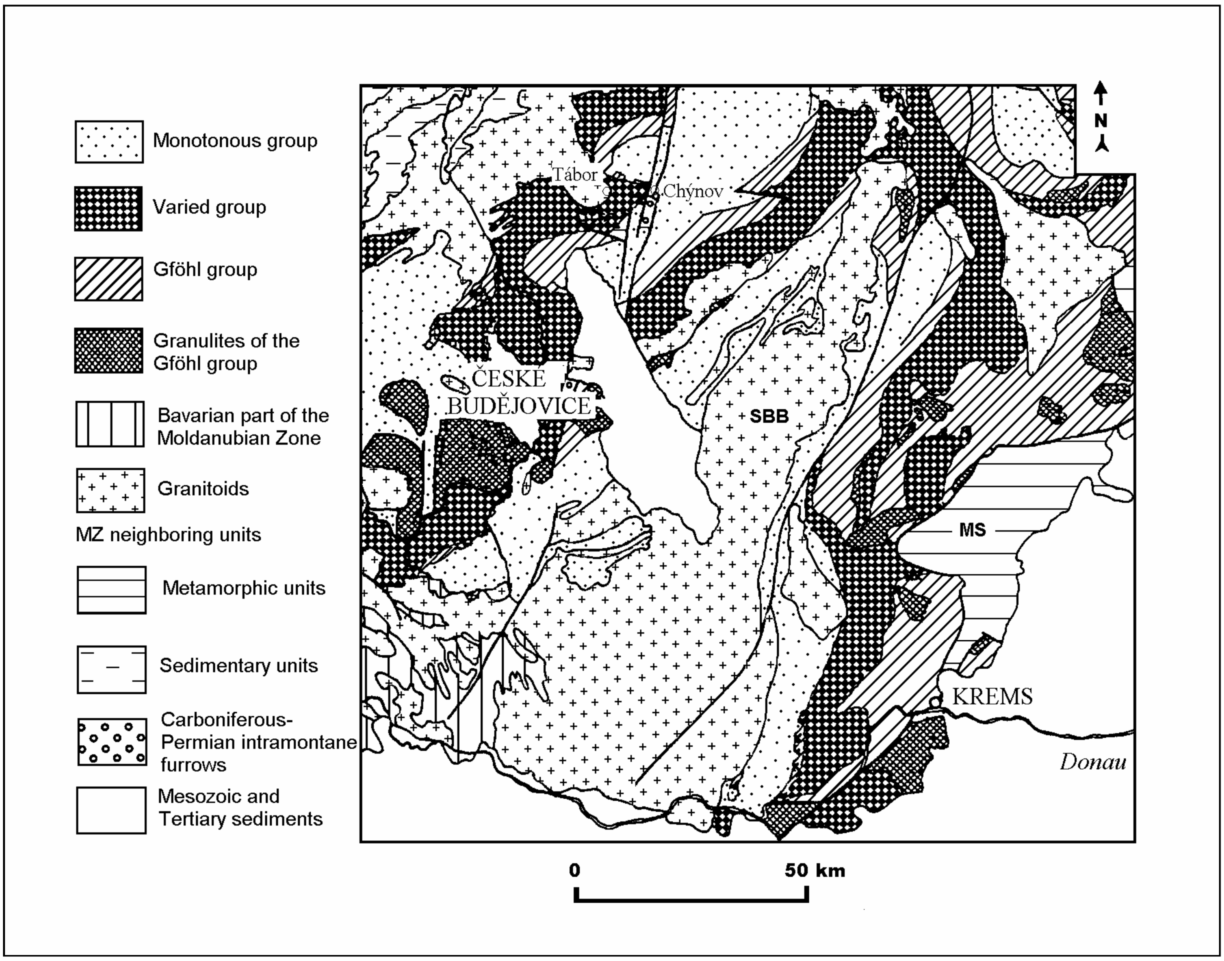

Fig. 1 Geological map of the southern part of the Moldanubian Zone (after Fiala, 1995; modified by author).

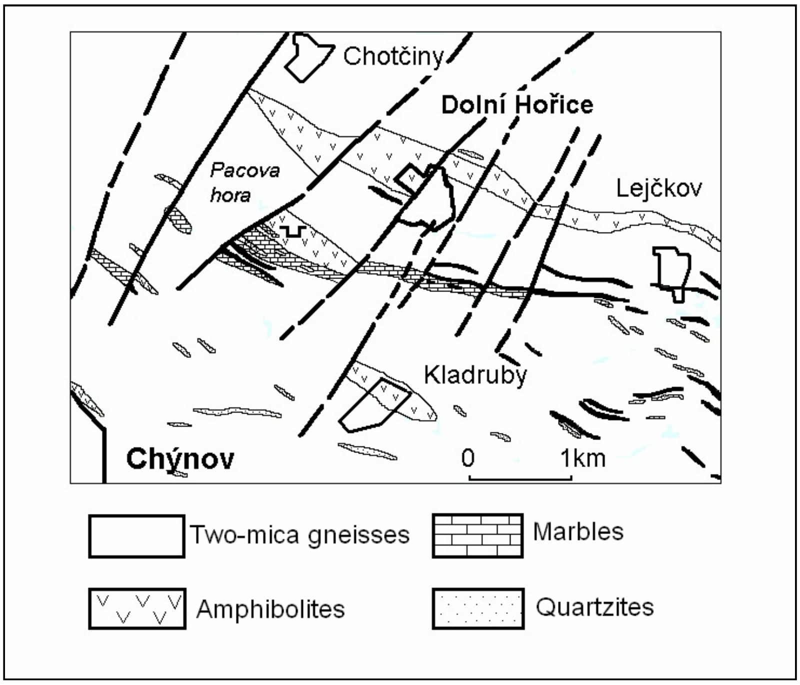

Fig. 2 Geological map of the Chýnov area. After Geological map 1:50,000 published by the Czech Geological Survey, sheet Tábor (Novák et al. ,1994; slightly modified by author).

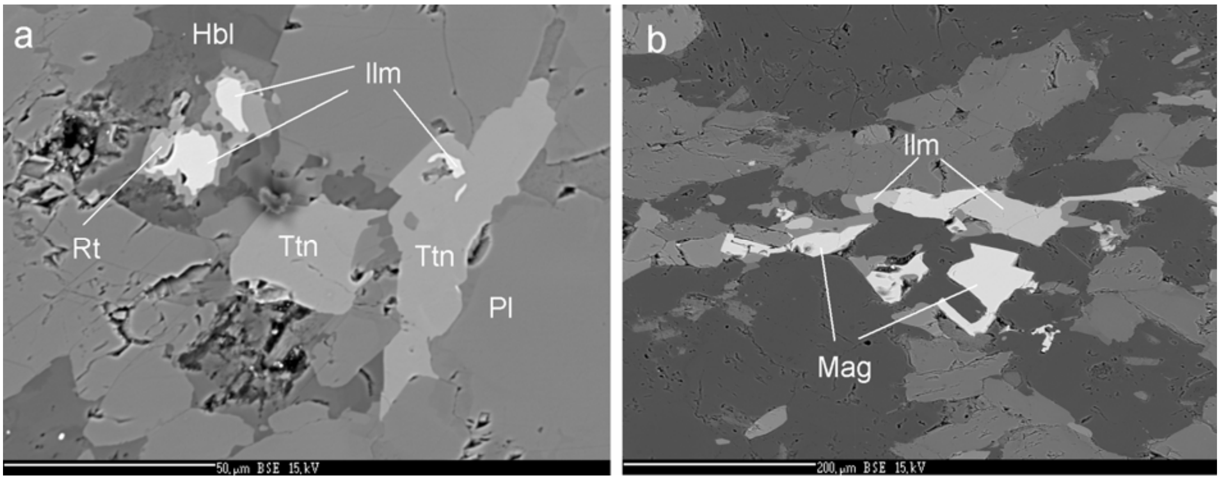

Fig. 3 a) Titanite reactions rims surrounding ilmenite, sample R-1581, Ttn – titanite, Ilm – ilmenite, Rt – rutile, Hbl – hornblende, Pl – plagioclase.
b) Ilmenite and magnetite mineral pairs in amphibolite, sample R-1581, Ilm – ilmenite, Mag – magnetite.

Janoušek et al., 1997). Amphibolites are accompanied by intercalations of calc-silicate rocks, quartzites and graphitic paragneisses. This area is characterized by the occurrence of plagioclase amphibolites, locally with a variable amount of carbonates (up to 10 vol. %).

Metamorphic series in the Chýnov area exhibit two to three successive deformation and recrystalization events in the predominant conditions of amphibolite facies (Vrána, 1979). The oldest MP/MT metamorphic phase is represented by an early kyanite-zoisite assemblage in Al-rich bands in marble and by presence of garnet bearing gneisses with garnet complex zoning pattern (Vrána, 1992). The main Variscan metamorphic phase is represented by assemblage amphibole + plagioclase ± pyroxene. The last metamorphic stage is characterised by cooling and by evolution of Late Variscan extension regime. In examined amphibolites is this stage characterised by occurrence of carbonate veinlets and chloritization of amphibole.

ANALYTICAL METHODS

Titanite and Fe-Ti-oxides analyses were performed on a CAMECA SX-100 electron microprobe employing the PAP matrix correction program (Pouchou and Pichoir, 1985) at the Institute of Geology of the Academy of Sciences of the Czech Republic. The operating conditions were 15 kV acceleration voltage, 15 nA beam current, and 2 µm beam diameter. Counting times on the peaks were 10-30 seconds depending on the element. Background counts were measured in each case in half the time for peak measurement on both sides of the peak. Calibrations were done using standard sets from SPI. Standards included jadeite (Na, Al), diopside (Si, Ca, Mg), rutile (Ti), hematite (Fe), spinel (Mn), crockoite (Cr), willemite (Zn) and apatite (F). Detection limits for these elements are as follows: F 0.09-0.15 wt.%, other elements 0.03-0.20 wt.%. Mineral formulae were recalculated using the Minpet 2.0 software.

PETROGRAPHY AND TEXTURAL RELATIONS

Amphibolites sampled in the abandoned quarry on the northwestern slope of Pacova hora Hill (samples R-1579 and R-1581) (Fig. 2) are formed by amphibole (50-70 vol.%) and plagioclase (20-40 vol.%). Clinopyroxene was found only occasionally in sample R-1579. Quartz, ilmenite, magnetite, titanite, rutile, apatite and sulphides (pyrite, chalcopyrite) are accessories. The high amount of ilmenite (up to 10 vol.%) and ilmenite rimmed by titanite and/or association of ilmenite with magnetite are the other significant features of these sampled amphibolites.

Titanite, together with Fe-Ti oxides (magnetite, ilmenite), usually forms subhedral grains and/or grain aggregates in amphibolites. Some thin sections of these amphibolites revealed relatively complex reaction rims of titanite around ilmenite. The titanite rims are nearly totally surrounded by plagioclase and/or are included in larger amphibole grains. Rutile inclusions are also present in some titanite grains (Fig. 3a). The magnetite-ilmenite intergrowths in the same rock form partly planar aggregates formed also by subhedral grains of ilmenite and magnetite. The majority of magnetite and ilmenite grains are surrounded by plagioclase and/or formed at the boundary between plagioclase and amphibole (Fig. 3b).

Table 1 Representative analyses of titanite (wt.%)

Sample	R-1579-5	R-1579-6	R-1579-10	R-1579-25	R-1579-31	R-1581-21
SiO_2	30.96	30.53	30.34	30.12	30.32	30.08
TiO_2	38.38	38.20	38.91	39.71	39.59	39.25
Al_2O_3	1.51	1.46	0.93	0.88	0.94	0.88
Fe_2O_3	0.60	0.68	0.70	1.04	0.84	1.23
MnO	0.01	0.04	0.09	0.19	0.06	0.09
CaO	28.03	27.87	28.10	28.09	28.04	27.68
Na_2O	0.01	0.00	0.03	0.03	0.03	0.03
F	0.48	0.52	0.08	0.20	0.14	0.23
O=F	0.14	0.16	0.02	0.06	0.04	0.07
Total	99.84	99.14	99.16	100.20	99.92	99.40
Si	1.000	1.000	1.000	1.000	1.000	1.000
Ti	0.932	0.941	0.965	0.992	0.982	0.982
Al	0.057	0.056	0.036	0.035	0.037	0.034
Fe^{3+}	0.015	0.017	0.017	0.026	0.021	0.031
Mn	0.000	0.001	0.003	0.005	0.002	0.003
Ca	0.970	0.978	0.992	0.999	0.991	0.986
Na	0.001	0.000	0.002	0.002	0.002	0.002
F	0.049	0.054	0.008	0.021	0.015	0.024
OH	0.023	0.019	0.045	0.040	0.043	0.041
Σ cations	3.047	3.066	3.068	3.120	3.093	3.103
X(Ttn)	0.928	0.928	0.948	0.942	0.945	0.938
X(Al,Fe^{3+}-F)	0.049	0.053	0.008	0.020	0.014	0.023
X(Al,Fe^{3+}-OH)	0.023	0.019	0.044	0.038	0.041	0.039

Formulae calculated on the basis of 1 Si; OH – Al+Fe^{3+}-F; Ttn: titanite, Al,Fe^{3+}-F: Al,Fe^{3+}-F titanite; Al,Fe^{3+}-OH: Al,Fe^{3+}-OH titanite.

MINERAL CHEMISTRY

Titanite

The most common substitution in natural titanite is the substitution of Al and Fe^{3+} for Ti, along with the coupled substitution of (Al, Fe^{3+}) + (F,OH) ⇔ Ti + O (Franz and Spear, 1985; Enami et al., 1993; Troitzsch and Ellis, 2002; Tropper et al., 2002; Harlov et al., 2006). The composition of titanite from the examined amphibolites ranges from 90 to 95 mol.% titanite end-member (Table 1). The Al and Fe contents range from 0.03 to 0.06 atoms per formula unit (a.p.f.u.) and 0.02 to 0.04 a.p.f.u., respectively, together with small amounts of F (Fig. 4). The Fe content in titanites which form reaction rims around ilmenite is low, about 0.02 a.p.f.u. Titanite also shows some Al+Fe^{3+} excess over F, which indicates the presence of the above mentioned Al, Fe^{3+}-involving substitution. The Fe^{3+} amounts are slightly lower than the Al contents, as shown in Table 1. Calculation of OH contents allows the estimation of the (Al+Fe^{3+})-OH titanite component, which ranges from 2 to 5 mol.%. The contents of OH-titanite in reaction rims of ilmenite are about 4 mol.%. The contents of (Al+Fe^{3+})-F component are more variable than the values of OH-titanite, and range from 1 to 8 mol.%. On average, the (Al+Fe^{3+})-F component slightly exceeds the (Al+Fe^{3+})-OH component.

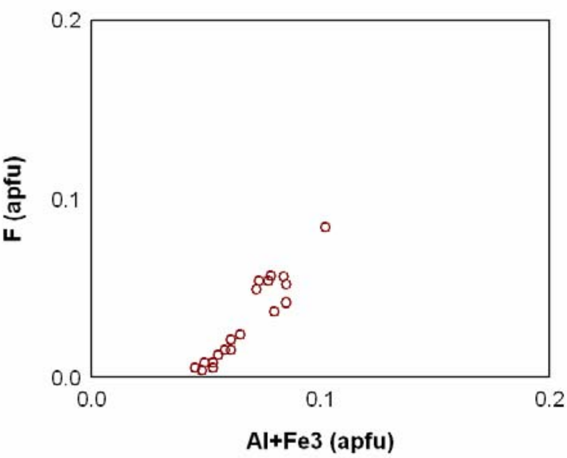

Fig. 4 Chemical variations in titanites from the sampled amphibolites of Pacova hora Hill (Chýnov area).

Table 2 Representative analyses of ilmenite (wt.%).

Sample	R-1579-23	R-1579-24	R-1579-30	R-1581-5	R-1581-6	R-1581-12
SiO_2	0.09	0.09	0.04	0.05	0.05	0.07
TiO_2	52.23	52.56	52.65	50.31	50.86	50.42
Cr_2O_3	0.00	0.00	0.07	0.02	0.00	0.10
Fe_2O_3	0.00	0.00	0.00	4.80	3.90	4.20
FeO	42.40	42.30	42.30	43.00	43.80	43.50
MnO	3.75	4.06	3.72	1.73	1.68	1.68
MgO	0.22	0.18	0.27	0.14	0.14	0.11
CaO	0.40	0.54	0.71	0.19	0.06	0.01
ZnO	0.00	0.00	0.00	0.07	0.05	0.00
Total	99.09	99.73	99.76	100.31	100.54	100.09
Si	0.002	0.002	0.001	0.001	0.001	0.002
Ti	0.997	0.997	0.998	0.953	0.961	0.958
Cr	0.000	0.000	0.001	0.000	0.000	0.002
Fe^{3+}	0.000	0.000	0.000	0.091	0.074	0.080
Fe^{2+}	0.900	0.892	0.891	0.906	0.920	0.918
Mn	0.081	0.087	0.079	0.037	0.036	0.036
Mg	0.008	0.007	0.010	0.005	0.005	0.004
Ca	0.011	0.015	0.019	0.005	0.002	0.000
Zn	0.000	0.000	0.000	0.001	0.001	0.000
X(Hm)	0.000	0.000	0.000	0.041	0.036	0.039
X(Ilm)	0.911	0.906	0.910	0.918	0.925	0.922
X(Gk)	0.008	0.007	0.010	0.005	0.005	0.004
X(XPPh)	0.081	0.087	0.080	0.036	0.034	0.035

Hm – hematite, Ilm – ilmenite, Gk – geikielite, Pph – pyrophanite. Recalculations of FeO_t using the method of Carmichael (1967).

Ilmenite

Ilmenite is a widespread Fe-Ti oxide in magmatic rocks as well as in regionally metamorphosed rocks, in both metapelites and metabasites (e.g., Carmichael, 1967; Thompson, 1972; Neumann, 1974; Rumble, 1976; Ghent et al., 1983; Ghent and Stout, 1984). Ilmenites represent a ternary solid solution of $FeTiO_3$, $MnTiO_3$, $MgTiO_3$ and Fe_2O_3. Some metamorphic ilmenites contain also appreciable amounts of Al_2O_3, CaO and Cr_2O_3 (Rumble, 1976). Ilmenites from amphibolites of the Chýnov area contain MnO as the dominant admixture with lesser amount of CaO, Al_2O_3, MgO and Cr_2O_3. The examined ilmenites typically have $FeTiO_3$ amounts above 96 mol.%, only some ilmenite grains are partly enriched in the $MnTiO_3$ (pyrophanite) component in amounts reaching 9 mol.% in some samples (Table 2). The contents of Fe_2O_3 in ilmenites are usually negligible, only some analysed magnetites have a low amount of the hematite component (2-5 mol.%).

Magnetite

The encountered natural magnetite is represented by a solid solution of Fe_3O_4 (magnetite) and Fe_2TiO_4 (ulvöspinel). The titaniferous magnetite is typical rather for magmatic than metamorphic rocks (e.g., Buddington and Lindsley, 1964; Czamanske and Mihálik, 1972; Neumann, 1974; Harlov, 2000). In metamorphic rocks, the ulvöspinel component is usually totally exsolved, and titaniferous magnetite is oxidized to an admixture of Ti-low magnetite and ilmenite.

In amphibolites from the Chýnov area, intergrowths of individual magnetite grains with ilmenite grains were found. The contents of the magnetite component in the analysed grains are very high, 99 mol.%, with negligible amounts of the ulvöspinel component (0.1-1 mol.%). The contents of other admixtures (Cr_2O_3, MnO, MgO, CaO and ZnO) in the examined magnetites is also very low (Table 3).

OXYGEN BAROMETRY

The oxygen barometry is based on the oxidation reaction between the magnetite component in the titaniferous magnetite and the hematite component in the ilmenite. Calculation of oxygen fugacity employed the QUIlF software developed by Andersen et al. (1993). Oxygen fugacities for the same ilmenite-magnetite mineral pairs range from $\log_{10}fO_2$ -26.0 to -27.5.

Table 3 Representative analyses of magnetite (wt.%)

Sample	R-1581-1	R-1581-3	R-1581-19
SiO_2	0.10	0.12	0.02
TiO_2	0.45	0.11	0.39
Cr_2O_3	0.05	0.06	0.04
Fe_2O_3	68.10	67.80	31.20
FeO	31.30	30.70	68.30
MnO	0.05	0.09	0.07
MgO	0.00	0.00	0.01
CaO	0.21	0.19	0.13
ZnO	0.08	0.01	0.00
Total	100.34	99.08	100.16
Si	0.004	0.005	0.001
Ti	0.013	0.003	0.011
Cr	0.001	0.002	0.001
Fe^{3+}	1.965	1.981	1.976
Fe^{2+}	1.004	0.997	1.003
Mn	0.002	0.003	0.002
Mg	0.000	0.000	0.001
Ca	0.009	0.008	0.005
Zn	0.002	0.000	0.000
X(Usp)	0.012	0.002	0.010
X(Mt)	0.988	0.998	0.990

Usp – ulvöspinel, Mt – magnetite. Recalculations of FeO_t using the method of Carmichael (1967).

DISCUSSION AND CONCLUSIONS

Oxide and titanite textures

The lack of ilmenite trellis structures in individual magnetite grains coupled with a low Ti content (TiO_2 0.1-0.5 wt.%) for all magnetite analyses in amphibolites from the Chýnov area would imply that the magnetite most likely never possessed a significant ulvöspinel component. In thin sections, the apparent reaction rims of titanite on ilmenite grains occur in a direct association with plagioclase and amphibole in amphibolites, thuis representing another sign of variable oxygen fugacity or H_2O activity during the Moldanubian metamorphism. Phase relations among titanite, ilmenite, amphibole and clinopyroxene (e.g., Xirouchakis and Lindsley, 1998; Frost et al., 2000b; Harlov and Hansen, 2005) and the lack of coexisting clinopyroxene at places with titanite rims around ilmenite imply that these parts of rocks should be characterized by either higher oxygen or H_2O fugacities compared with the clinopyroxene and ilmenite assemblages. The titanite reactions rims at these places probably originated by the following reactions:

$$\text{6 Fe-actinolite} + \text{12 Ilmenite} + 7\,O_2 = \text{12 Titanite} + \text{14 Magnetite} + \text{36 Quartz} + 6\,H_2O.$$

Such reaction implies that the water fugacity must also increase with increasing oxygen fugacity. Regardless of the mode of titanite formation, as TiO_2 is removed from ilmenite to form titanite, the residual ilmenite becomes enriched in the hematite component. However, the residual ilmenite inclusions in distinctly larger titanite (Fig. 3a) show no enrichment in hematite component. On the other hand, parts of some titanite rims also contain very small rutile inclusions (Fig. 3a). The assemblage of titanite, rutile and ilmenite probably implies distinctly high oxygen fugacity. Ilmenites rimmed by titanite similar to amphibolites from the Chýnov area in their texture, have been reported from felsic orthogneisses (Tropper and Hoinkes, 1996; Harlov and Förster, 2002; Harlov and Hansen, 2005; Harlov et al., 2006), amphibole-bearing eclogites (Liu et al., 1998) as well as amphibole-bearing granitic rocks (Frost et al., 2000a; Zachovalová et al., 2002). In assemblages with abundant clinopyroxene, other reactions have been also suggested for the origin of titanite rims around ilmenite. These are based either on rehydration:

$$\text{7 Clinopyroxene} + \text{3 Ilmenite} + \text{5 Quartz} + 2\,H_2O = \text{2 Amphibole} + \text{3 Titanite}$$

or oxidation:

$$\text{3 Clinopyroxene} + \text{3 Ilmenite} + O_2 = \text{3 titanite} + \text{2 magnetite} + \text{3 Quartz.}$$

Both these reactions involve clinopyroxene (e.g., Xirouchakis et al., 2001 a, b; Harlov and Hansen, 2005). The low contents of clinopyroxene and its restitic character in thin sections, together with the occurrence of titanite rims in the examined Moldanubian amphibolites cannot exclude the occurrence of the above mentioned reactions.

Titanite stability as a function of fH_2O and fO_2

The occurrence of ilmenite rimmed by titanite in amphibolites from the Chýnov area, together with the presence of the ilmenite+magnetite assemblage in the same area show that a higher fluid and oxygen activity was very feasible. The chemical composition of the examined titanites shows that the substitution (Al, Fe^{3+}) + F ⇔ Ti + O as well the substitution (Al, Fe^{3+}) + OH ⇔ Ti + O were present. In addition to the Al-F substitution, also the Al-OH substitution is highly important for natural titanites (e.g., Enami et al., 1993; Harlov et al., 2006). According to the study of Enami et al. (1993), the presence of the Al-OH substitution is highly significant for LP-LT metamorphism, but Markl and Piazolo (1999) have shown that the chemical composition of Al-bearing titanite is a more complex function of several variables. Data from amphibolite facies conditions in the Moldanubian Zone of the Bohemian Massif indicate that both Al-F-bearing titanite and Al-OH-bearing titanite form a stable association with hornblende. This assemblage is highly significant for high fO_2 and fH_2O (e.g., Harlov et al., 2006). Negligible hematite contents in ilmenite suggest that, in most samples, Fe (as Fe^{2+}) was preferentially partitioned in coexisting ferromagnesian minerals such as amphibole or garnet.

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