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Geochemical correlation of metabasic rocks from Central and East Rhodopes, Bulgaria

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Abstract. Metaigneous rocks, including metagabbros and amphibolites, are well known elements of the Variegated Formations from Central and East Rhodopes. A comparison between the geochemical features of these rocks from Madan-Davidkovo Antiform (Central Rhodopes) along with the Avren Synform, and Bela Reka Antiform (East Rhodopes) is the aim of this contribution. The chemical composition of metabasic rocks from both regions, e.g., high MgO, and low high field strength elements (*HFSE*) contents, indicates boninite and arc tholeiite affinities. Such affinities are also reflected by the values of specific ratios such as CaO/Al_2O_3, CaO/TiO_2, Al_2O_3/TiO_2, La/Sm. Plotted on discrimination diagrams, the metabasic rocks from Variegated Formations in both regions fall mainly in the fields of Phanerozoic boninites and arc tholeiites. Three types of *REE* patterns are observed in the metabasites from these regions: U-shaped, flat to *LREE* enriched pattern, and *LREE* depleted pattern found in the rocks from Avren Synform only. Regardless of the existence of diverse *REE* patterns, the La/Sm ratios perfectly overlap the range of the same ratios for many Phanerozoic boninites and low-Ti tholeiites associated with Cambrian boninitic rocks.

The compositional similarity of the correlated metabasic rocks (boninite-like characteristics) along with the features of the Variegated Formations from both regions, may be used as an important key when comparing their general geological structure and geodynamic evolution.

Key words: boninite, island arc tholeiite, rare earth and trace elements, Rhodopes

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Лилан-Ана Даиева, Иван Хайдутов, Стефка Приставова. Геохимична корелация на метабазични скали от Централните и Източни Родопи, България

Резюме. Добре известно е присъствието на метаморфозирани базични магмени скали (метагабра и амфиболити) като елемент на пъстрите формации от Централните и Източни Родопи. В тази публикация е направено сравнение на геохимичните особености на тези скали от Мадан-Давидковската антиформа (Централни Родопи), Авренската синформа и Белоречката антиформа (Източни Родопи). Химичният състав на метабазитите от двата района, високо съдържание на MgO и ниско - на елементи с висока сила на полето (*HFSE*), сочи сходството им с бонинити и островнодъгови толеити. Стойностите на специфични отношения като CaO/Al_2O_3, CaO/TiO_2, Al_2O_3/TiO_2 и La/Sm потвърждават това сходство. Проектирани върху дискриминационни диаграми, метабазитите от пъстрите свити на двата района попадат главно в полетата на фанерозойските бонинити и островнодъгови толеити. Наблюдавани са три вида хондритнормирано разпределение на

REE в метабазитите от двата района: U-видно, равномерно до слабо обогатено на леки редкоземни елементи и обеднено на леки редкоземни елементи разпределение (наблюдавано само в скалите от Авренската синформа). Независимо от съществуването на различни по вид хондритнормирани разпределения на *REE*, стойностите на отношението La/Sm перфектно съвпада със стойностите на същото отношение в много фанерозойски бонинитови и ниско-Ti толеити, свързани с камбрийски бонинитови скали.

Геохимичната близост на корелираните метабазити (бонинитов тип) заедно с особеностите на пъстрите свити от двата района може да бъде използвано като важен аргумент за изясняване на геоложкия строеж и геодинамична еволюция на последните.

Introduction

Boninitic rocks are characterized by high variation in composition, from high-Ca low-Si types, to low-Ca high-Si types. Based on petrological observations (Beccaluva, Serri, 1988) and analysis of phase equilibrium during melting (van der Laan et al., 1989), a continuum from basalt to low-Ca boninite is supposed also to exist for this rock type. Boninites are widely recognized in fore arc basement sequences in the Bonin-Marianas region of the Western Pacific (Hickey, Frey, 1982; Reagan, Meijer, 1984; Blomer, Hawkins, 1987; Pearce et al., 1992; Taylor et al., 1994). The occurrence of boninitic rocks is reported in several ophiolite complexes such as Koh (Meffre et al., 1996), Betts Cove (Coish, 1989), as well as the Lower Proterozoic Trans-Hudson orogen (Wyman, 1999).

The presence of metaigneous rocks, including metagabbros and amphibolites, in the high-grade metamorphic rocks from the Central and East Rhodopes, is well documented in the geological literature (Kozhoukharov, 1965; Kozhoukharova, 1996). Special attention is paid to the petrological and mineralogical features of these metabasic rocks, as well as to the metamorphic conditions of their recrystallization (Toprakchieva, 1963; Pristavova 1996; Haydoutov et al., 2004). Recently, several authors studied the geochemistry of metabasic rocks from the Central and East Rhodopes (Zakariadze et. al., 1993; Daieva, Pristavova, 1998; Bazylev et. al., 1999; Haydoutov et al., 2004) and demonstrated their boninite-like affinity. The geochemical correlation of boninite-like rocks of the Variegated Formations from both regions is the aim of this study. This correlation is of great value, especially for comparing the general geological features of the investigated areas.

Geological position and petrographic characteristics

The studied metabasites (metagabbros) from the Central Rhodopes crop out in the East periphery of the Madan-Davidkovo Antiform (MDA) (Fig. 1). They occur as several small bodies, hosted in metamorphic rocks of the Vishnevo Formation (Ardino Group), which is composed of gneisses, affected by granitization and migmatization, amphibolites, and schists (Kozhukharov, 1988). According to the lithotectonic subdivision of the metamorphic rocks from the Central Rhodopes (Sarov et al., 2003) these rocks are related to the Arda lithotectonic units, built up by diatexite-type migmatites and anatectite granites with Variscan age of the protholith (Peytcheva et al., 2003). The metagabbro is included as small lenses among amphibolites in these granitoid lithologies. In the central parts of their bodies, these rocks are with well-preserved igneous texture and mineral composition - Mg-rich olivine, plagioclase, ortho- and clinopyroxene, high-Ti phlogopite, and spinel (Pristavova, 1996). Corona (druzite-like) textures are established in these rocks. Magmatic minerals and textures gradually diminish towards the periphery of the metabasite bodies. Pristavova (1996) determine magmatic temperature for the metabasites of about 1160-1170°C, and pressure about 7 kbar.

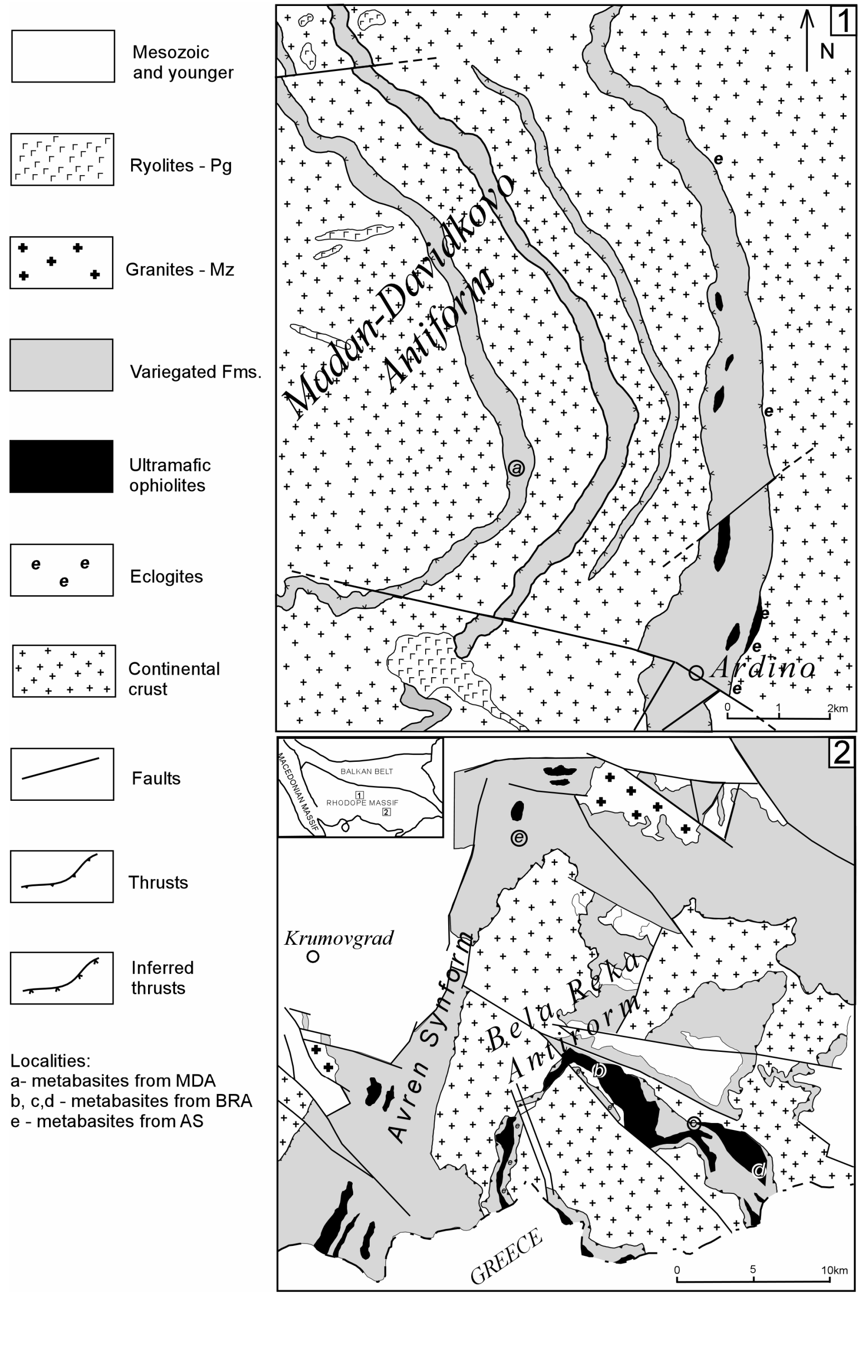

Fig. 1. Simplified geological maps: 1 - the east part of Madan-Davidkovo Antiform (MDA); 2 - Avren Synform (AS) and Biala Reka Antiform (BRA) showing the main tectono-stratigraphic units. Compiled after Kozhouharov (1988) and Haydutov et al. (2004) with additions of the authors

During subsequent metamorphism, the igneous minerals were replaced by garnet, amphibole, plagioclase, titanite and chlorite. The *P/T* condition of this metamorphic process in the hosted amphibolites is determined at about 550-770°C and 6 kbar (Pristavova, 1996).

The metabasites from the East Rhodopes (Haydoutov et al., 2004) occur as layers and slices that are interlayerd with metasediments (in the Avren Synform), or as intrusive bodies intersecting the ultramafic fragments of the ophiolitic units (mainly in the Bela Reka Antiform) (Fig.1). They are fine- to medium-grained, mesocratic to melanocratic rocks, massive in the internal parts of the bodies, and usually foliated in their external parts, with rare relics of ophitic textures preserved. The rock forming minerals in these metabasites are amphibole + plagioclase + quartz + epidote ± garnet ± chlorite. Accessory phases include titanite, apatite, rutile, magnetite, and zircon. The calculated temperatures of 630-520°C and pressure of 6-2 kbar indicate moderate amphibolite facies metamorphism (Haydoutov et al., 2004). The above mentioned meta-sediments, interlayerd with the metabasic rocks, include metaterrigenous rocks (meta-grauwaks, metapsamites), metapelites and marbles. They are typical for an island arc environment (Haydoutov et al., 2004). Their intercalation with the considered here meta-basics shows that this assemblage was formed in such an environment. The island arc is formed over the ophiolites (ocean crust) evidenced by the cross-cutting relations, observed mainly in Bela Reka Antiform.

Geochemical features of the metabasic rocks

The most striking features of boninites, as arc-related rocks, are their high MgO (>6%) and low TiO_2, Zr and *HREE* contents. Characteristic for the metabasites from the Madan-Davidkovo Antiform are variable MgO contents (reaching 24.2%, Table 1), TiO_2 ~0.60% and Zr ~40 ppm. The contents of these elements in the metabasites from East Rhodopes are as follow: average MgO~6.70%, TiO_2~0.76% and Zr~60 ppm. Although the observed various MgO contents in the studied metabasites, their Mg-number range between 55 and 80 and show that most of them are close to the primitive mantle derived magmas (Bloomer, Hawkins, 1987). This fact is supported by parallel Ni-, Cr- and Mg-enrichment in both regions, better pronounced in the rocks from MDA.

Plotted on AFM diagram the samples from the Central and East Rhodopes fall predominantly in MORB-field, but using the TiO_2 *vs*. Zr and Cr *vs*. Y (the last not shown here) plots of Pearce (1980) they fall in the island arc field (Fig. 2). Because of their lower TiO_2 content some of the rocks from MDA fall close under the lower margin of VAB field (Fig. 2). For this reason on the discrimination diagram of Mullen (1983) MnOx10-TiO_2-P_2O_5x10 (not shown here) the investigated metabasites are divided in two groups. These with lower TiO_2 content (from MDA and Avren Synform) occupy the boninite field and the rest with more elevated TiO_2 content (from Bela Reka Antiform) occupy the island arc tholeiite field. The close similarity to the

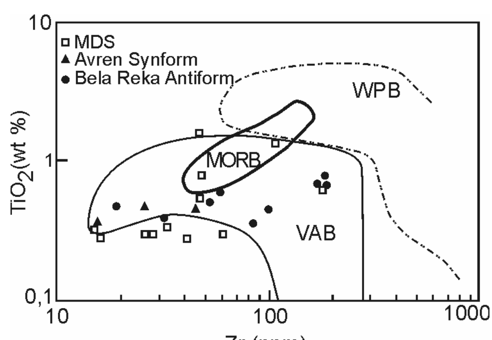

Fig. 2. TiO_2 *vs*. Zr discrimination diagram for boninite-like rocks from Central- and East Rhodopes: VAB - volcanic arc basalts, WPB - within plate basalts, MORB - mid ocean ridge basalts (after Pearce, 1980)

Cenosoic boninites is clearly seen on $Mg^{\#}$ *vs.* TiO_2 diagram (Pearce et al., 1992). The investigated metabasites from MDA and the most primitive rocks from Avren Synform reside in the area of fields overlapping between Zambales ophiolite boninites and MORB, Mariana Trench and DSPD Site 786 boninites (Fig. 3). The metabasic rocks from Bela Reka Antiform plots in the fields of DSPD Site 458 and Zambales ophiolite boninites. Characteristics such as CaO/TiO_2, Al_2O_3/TiO_2, Ti/Zr, Ti/Y, Zr/Y ratios are transitional between boninitic and island arc tholeiitic rocks. According to the classification of Crawford et al. (1989) the metabasites from both regions fall into the low-Ca boninite group with $CaO/Al_2O_3<0.75$.

An important characteristic of boninitic and boninite-like rocks is their chondrite- normalized *REE* pattern. Three types of *REE* patterns are observed in both studied regions:

- A U-shaped pattern, found in many Cenosoic boninites from Bonin Islands (Taylor et al., 1994), Cape Vogel and DSDP 458 (Pearce et al., 1992), as well as Appalachian ophiolites (Coish, 1989) is typical for samples from Madan-Davidkovo Antiform, Avren Synform and Bela Reka Antiform (Fig. 4a, Table1).

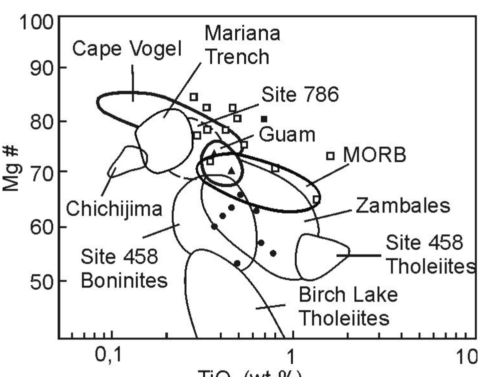

Fig. 3. $Mg^{\#}$ *vs.* TiO_2 discrimination diagram for the rocks studied (fields after Pearce et al., 1992 and Wyman, 1999). For symbols see Fig. 2

This pattern is characterized by light- and heavy *REE* enrichment (La_N/Sm_N 1.5-3.1, Gd_N/Yb_N 0.5-0.8) and positive Eu anomaly (Eu' 1.2-1.9).

- A nearly flat to slightly *LREE* enriched patterns, observed in Mariana trench boninites (Bloomer, Hawkins, 1987) is typical for most

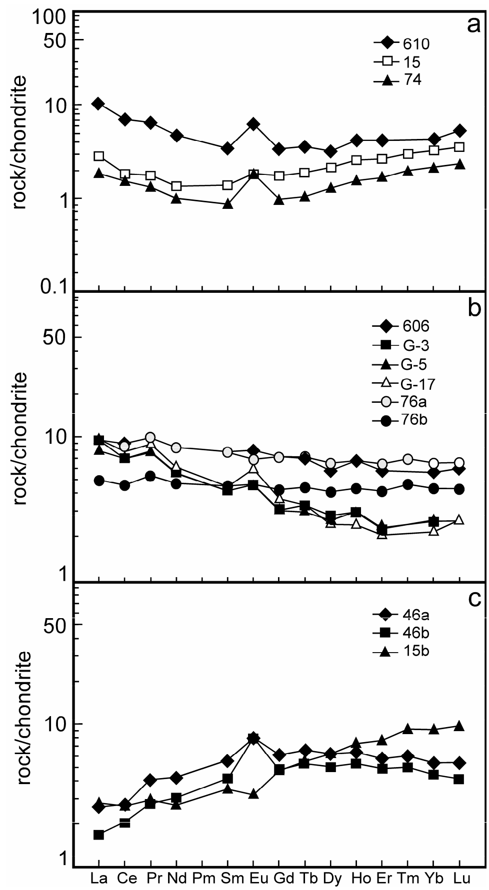

Fig. 4. Chondrite-normalized *REE* pattern for boninite-like rocks from Central and East Rhodopes: a) U-shaped *REE* pattern in rocks from Madan-Davidkovo Antiform, Avren Synform, Bela Reka Antiform; b) flat to *LREE* enriched pattern in rocks from same structures; c) *LREE* depleted pattern in rocks from Avren Synform. For symbols see Fig. 2 and Table 1

Table 1. *Chemical composition of selected metabasic rocks from Central and East Rhodopes*

	Madan-Davidkovo Antiform					Avren Synform				Bela Reka Antiform		
wt.%	G-3[1]	G-5[1]	G-17[1]	606[2]	610[2]	46a[3]	46b[3]	15[3]	15b[3]	74[3]	76a[3]	76b[3]
SiO_2	43.71	44.58	45.14	50.38	47.47	48.22	47.55	51.63	57.67	45.73	53.37	50.50
TiO_2	0.30	0.32	0.30	0.77	0.34	0.47	0.36	0.01	0.45	0.48	0.61	0.40
Al_2O_3	11.64	18.09	17.50	16.84	18.10	16.55	17.60	17.11	15.31	20.65	15.08	17.07
Fe_2O_3	2.95	1.10	9.20	n.d.	n.d.	2.20	1.90	3.20	4.90	3.24	3.27	3.61
FeO	7.19	6.06	n.d.	6.44	8.21	4.73	4.20	5.47	6.60	6.16	4.76	4.74
MnO	0.29	0.11	0.15	0.12	0.12	0.11	0.11	0.13	0.18	0.07	0.12	0.09
MgO	24.18	14.56	13.36	9.06	12.65	9.07	9.80	7.15	3.05	5.87	7.78	7.70
CaO	7.47	10.72	10.06	12.35	10.03	12.69	12.98	11.12	7.42	12.34	7.40	8.58
Na_2O	0.94	1.60	1.48	2.07	1.75	2.61	2.26	1.36	1.77	2.18	4.94	4.44
K_2O	0.22	0.21	0.29	0.36	0.15	0.30	0.30	0.24	0.33	0.20	0.08	0.15
P_2O_5	0.04	0.03	0.04	0.12	0.10	0.04	0.03	0.03	0.08	0.02	0.09	0.03
LOI	1.34	2.40	1.54	0.75	1.30	2.76	2.70	2.43	2.46	2.82	2.48	2.50
Mg#	85	79	73	73	72	71	75	60	33	54	64	63
ppm												
Rb	8	6	7	4	3	7	3	27	13	3	5	3
Ba	96	58	91	n.d.	n.d.	86	73	30	79	88	137	128
Sr	205	290	289	242	298	361	273	53	62	175	115	174
Zr	26	15	28	48	33	26	16	<7	<7	19	59	32
Y	11	11	10	18	14	12.8	10.4	5.9	15.9	3.3	14.1	8.8
Nb	0.81	0.88	13.41	n.d.	n.d.	n.d.	n.d.	<5*	n.d.	n.d.	n.d.	n.d.
La	3.13	2.74	3.25	3.16	3.37	0.88	0.55	0.64	0.94	0.62	3.11	1.66
Ce	6.19	6.05	6.89	8.01	6.10	2.38	1.75	1.60	2.35	1.34	7.55	4.02
Pr	0.90	0.89	1.04	1.10	0.72	0.46	0.31	0.20	0.34	0.15	1.12	0.60
Nd	3.62	3.40	4.04	5.40	3.00	2.68	1.92	0.85	1.73	0.64	5.35	2.95
Sm	0.87	0.89	0.95	1.62	0.70	1.12	0.85	0.28	0.72	0.18	1.60	0.93
Eu	0.36	0.47	0.47	0.63	0.48	0.63	0.61	0.14	0.25	0.14	0.54	0.36
Gd	0.88	0.88	1.06	2.00	0.93	1.69	1.34	0.45	1.32	0.27	2.01	1.20
Tb	0.16	0.15	0.16	0.34	0.17	0.31	0.25	0.09	0.27	0.05	0.34	0.21
Dy	0.99	0.93	0.90	2.00	1.10	2.13	1.73	0.74	2.15	0.45	2.26	1.44
Ho	0.21	0.21	0.18	0.48	0.29	0.45	0.37	0.18	0.52	0.11	0.48	0.31
Er	0.52	0.56	0.49	1.30	0.94	1.31	1.10	0.60	1.75	0.39	1.46	0.95
Tm	n.d.	n.d.	n.d.	n.d.	n.d.	0.18	0.15	0.09	0.28	0.06	0.21	0.14
Yb	0.52	0.55	0.46	1.15	0.94	1.18	0.98	0.72	2.02	0.48	1.45	0.97
Lu	0.01	0.09	0.09	0.21	0.20	0.18	0.14	0.12	0.33	0.08	0.22	0.15

[1)] after Daieva, Pristavova, 1998; [2)] after Zakariadze et al., 1993; [3)] after Haydoutov et al., 2004;
[*)]ICP-AES - N. Velitchkova; n.d.- not determined

samples from MDA and some from Bela Reka Antiform (Fig. 4b, Table 1). The former is characterrized by La_N/Sm_N 1.1-1.2, Gd_N/Yb_N 1.1-1.4 and no Eu anomaly. The slight *LREE* enriched patterns have slightly elevated values of La_N/Sm_N 2.0-2.3, Gd_N/Yb_N 1.3-1.9 and positive Eu anomaly.

- A pattern with pronounced *LREE* depletion (Fig. 4c) is encountered in the boninite-like rocks from Avren Synform (Table 1). This pattern is characterized with La_N/Sm_N 0.4-0.8, Gd_N/Yb_N 0.5-1.2 and varying from slightly negative (0.8) to positive (1.7) Eu anomaly. Light *REE* depleted patterns are reported for boninitic rocks from Marianas, New Caledonia and Lau Basin (Crawford et al., 1989; Cameron, 1989; Hawkins, 1995).

The positive Eu anomaly in the most of the studied metabasites is interpreted as being result of plagioclase accumulation.

Phanerozoic boninites typically define distinct fields on plots involving TiO_2, Zr and *REE*. Plotted on La/Sm *vs.* TiO_2 diagram (Wyman, 1999) the metabasites from MDA and East Rhodopes fall within the field of Cenosoic boninites, but two sample from Avren Synform fall in the field of low-Ti tholeiites associated with Cambrian boninitic rocks (Fig. 5). On a Zr/Sm *vs.* La/Sm diagram (Pearce et al., 1992, not shown here) the metabasic rocks from both regions are close to the fields defined by island arc tholeiitic rocks.

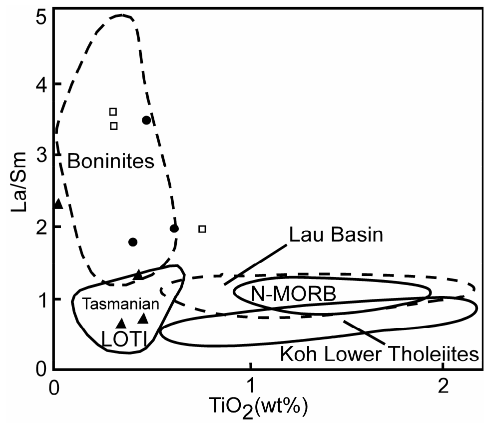

Fig. 5. La/Sm *vs.* TiO_2 discrimination diagram for boninite-like rocks from Central and East Rhodopes (fields after Wyman, 1999)

Discussion

The comparison between the metabasites of the Central and East Rhodopes clearly shows the compositional similarity of these rocks. Moreover, their position on the discrimination diagrams (Cr *vs.* Y, TiO_2 *vs.* Zr, $Mg^{\#}$ *vs.* TiO_2, Zr/Sm *vs.* La/Sm), and the values of specific ratios (e.g. CaO/Al_2O_3, CaO/TiO_2, Al_2O_3/TiO_2, La/Sm), demonstrate the similarity of the mentioned rocks to the Cenosoic boninitic and tholeiitic rocks. The observed diverse *REE* pattern is typical for many boninite sequences (Cameron, 1989; Meffre et al., 1996). The established slight *LREE*-enrichment in the metabasites from the Central Rhodopes can be regarded as a probable result of later alterations (Alpine metamorphism and migmatisation) which can not change their position in the fields of Cenosoic boninitic and tholeiitic rocks on the discrimination diagrams involving *REE*'s (see Fig. 5).

The most characteristic geochemical features of Cenosoic boninitic rocks are the negative Nb and Ti anomalies in their MORB-normalized trace element pattern and depletion of *HFSE* compared to *LILE* (Bloomer, Hawkins, 1987). This pattern is established for boninite-like rocks from Central Rhodopes (Daieva, Pristavova, 1998). New analytical data for Nb concentration in Bubino metagabbro (Table 1) shows the existence of negative Nb and Ti anomalies on Fig. 6. The same diagram displays depleted patterns at about 0.5-0.7xN-MORB (Sun, McDonough, 1989) for the MORB-normalized *HFSE* concen-trations. This trace elements pattern is observed in supra subduction zone boninites from Vardar zone (Marroni et al., 2004). Moreover, the comparison of boninite-like rocks from Central and East Rhodopes with Cenosoic boninitic rocks from LEG 125 (Bonin-Mariana Forearc, Pearce et al., 1992) and from Bonin Islands (Taylor et al., 1994), demonstrates the close similarity between them (Fig. 6).

The importance of the established above similarity between Central and East Rhodopes boninite-like rocks can be best appreciated when comparing the general structures of these areas.

East Rhodopes. The high-grade metamorphic basement in this region is formed by two units. The lower one, exposed in the cores of antiforms or domes – Bela Reka and Kessibir, consists of orthogneiss succession of continental affinity (Macheva, Kolcheva, 1992; Ricou et al., 1998; Haydoutov et al., 2004). The age of the protolith is Variscan (Peytcheva,von Quadt, 1995). The upper metamorphic unit, exposed mainly in the synform structures

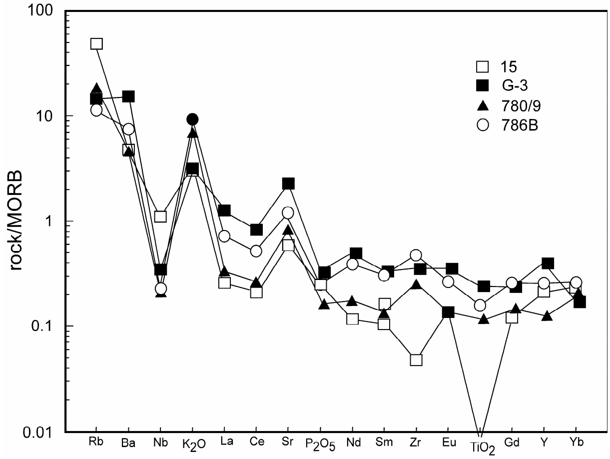

Fig. 6. Spidergrams of metagabbros G-3 (Madan-Davidkovo Antiform) and 15 (Avren Synform), compared to Cenozoic boninites of Bonin - Mariana Fore arc, 786B (Pearce et al., 1992) and 780/9 (Taylor et al., 1994). Data are normalized to the N-MORB composition (Sun, McDonough, 1989)

(e.g. Avren, Snezhina), and is formed by a double layered assemblage. The lower layer consists of fragments of oceanic crust, overlain by the interlayered metamafic and sedimentary components of the Variegated Formation. The protolith ages for these rocks are generally not known with exception of a single U-Pb zircon determination suggesting that the metaigneous rocks crystallized in the late Neoproterozoic (572±5 Ma, Carrigan et al., 2003). The contact between the Variscan continental crust and the double layered assemblage (ophiolites and Variegated Formation) is tectonic. In most of the studied localities, the contact shows features of a deep tectonic structure with internal tectonic imbrications.

The considered units were characterized as two different terranes. The lower terrane with continental affinities is named Bela Reka, while the upper one – is named Rhodopes. The upper terrane is of particular interest for this research. It is formed by ophiolites and rocks of primitive ensimatic island arc. This composition of the upper terrane is of crucial importance for understanding the general structure of the considered region. Obviously, its position on top of the continental crust (Bela Reka terrane) is result of a large scale thrusting, evidenced by the features of the contact between them. The time of this thrusting could be latest Variscan or later. This process could be connected with the Variscan collision from a general point of view.

Central Rhodopes. The studied metabasites are situated in the east periphery of the MDA. The high-grade metamorphic basement in this region is formed by two units analogous to the East Rhodopes. The lower one, exposed in the cores of structure is built of biotite and two-mica gneisses, migmatites and anatectite granites with Variscan age of the protholith (Peytcheva et al., 2004). The upper metamorphic unit, exposed in the East periphery of the structure is formed by the Variegated Formation, which contains ultramafic rocks (mainly in the lower part), amphibolites, high-Al mica schists and marbles. The protolith ages for these rocks are also generally not known. A single U-Pb zircon dating of eclogites of this region (600 Ma, Arkadakskiy et al., 2003) shows late Neoproterozoic age of their crystallization. The numerous sub-parallel layers of Variegated Formation, embedded among the rocks of the continental crust (Fig. 1), could be interpreted as traces of the tectonic imbrications along the contact between both units. Analogous tectonic position is suggested by Pristavova and Kozhoukharova (1999) investigated the structure position of Variegated Formation in this region. The unclear contact between the Variegated Formations and their basement formed mainly by metagranites in Central Rhodopes in comparison with the clear tectonic contact between these units in East Rhodopes (Haydoutov et al., 2004), is a problem. In the MDA this contact is probably influenced by the intense alpine metamorphic overprint.

Conclusions

Several conclusions could be drawn, as follows:

- The metabasic rocks from the Variegated Formation of both regions show similar boninite-like features. This similarity is the base for the correlation of both regions' general structures;
- The Central and East Rhodopes regions show features of comparable general geological structure. They are composed of a Variscian continental terrane at the base of the metamorphic profile. A completely different terrane is situated on top of this continental crust. This is the double-layered unit, composed of the ophiolite assemblage over-layered by the components of a primitive ensimatic island arc (Variegated Formation);
- In the East Rodopes the two terranes were indicated as Biala Reka and Rhodopes. For both compared regions, the Variscan terrane is named as "Low Rhodopes", and the upper - "Rhodopes" terrane. The position of the "Rhodopes" terrane thrusted above the "Low Rhodopes" terrane clearly shows the presence of a suture zone, indicated as "Rhodopes suture zone".

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