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Evidence of boninitic type magmatism in the Variegated Formations from the East Rhodope

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Abstract. The Variegated Formations of the eastern Rhodope Mountains are composed of alternating igneous rocks and sediments with a high-grade metamorphic overprint. Numerous ophiolitic slivers are associated with these formations. They comprise metamorphosed peridotites, ultramafic cumulates, and amphibolitized eclogites. The ophiolites (intensively dismembered) usually form the basement of the Variegated Formations.

The metaigneous rocks of the Variegated Formations occur either as layers interbedded with metasediments, or as intrusive bodies that intersect the ultramafic rocks. The principal phases in the metabasites are amphibole + plagioclase + quartz + epidote ± garnet ± chlorite. We calculate temperatures of 630°C to 520°C at pressures of 6-2 kbar, indicating moderate amphibolite facies metamorphism. Major rock forming minerals (amphibole, plagioclase, and garnet) exhibit zoning typical of retrograde *P-T* conditions.

The chemical composition of the studied metaigneous rocks indicates boninite and arc-tholeiite affinities. They include low Ti and Zr content and also the key ratios of CaO/TiO_2, Al_2O_3/TiO_2, Ti/Zr, Ti/Y and Zr/Y, all transitional between island arc tholeiites and boninites. Plotted on a variety of discrimination diagrams, the metabasic rocks of the Variegated Formations fall mainly in the fields of modern boninites and arc tholeiites. The chondrite-normalized *REE* patterns reveal the existence of two different trends: U-shaped *REE* patterns (for the majority of samples) and *LREE* depleted patterns. Regardless of the existence of these two trends, the $[La/Sm]_N$ ratios of the metabasites perfectly coincide with the same ratios for many Cenozoic boninite series. The metasedimentary rock types contain terrigeneous materials (metapsammites and quartzites) that frequently alternate with metapelites and marbles. The nature of this sedimentary package reflects its flysch character.

The clear boninitic and arc-tholeiite affinities of the igneous rocks, as well as the character of the sedimentary sequences, indicates that the Variegated Formations formed in an oceanic island-arc environment. The mentioned affinities of the meta-igneous rocks indicate an origin in an immature arc. The character of the Variegated Formations and its association with the dismembered ophiolite slivers shows the presence of a suture zone. The East Rhodope suture zone distinguishes the Variegated Formations from the rocks structurally below it, which consist of orthogneisses typical of continental crust. Existing U-Pb zircon data indicate that the orthogneisses are of Variscan age. New U-Pb zircon age data for the Variegated Formations suggest Late Neoproterozoic ages for some protoliths.

Key words: boninite, island arc, ophiolite, Neoproterozoic, Rhodope.

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Резюме. Пъстрите свити от Източните Родопи са изградени от алтерниращи магмени скали и седименти, засегнати от високостепенен метаморфизъм. Многобройни офиолитови блокове асоциират с пъстрите свити. Блоковете са изградени от метоморфозирани перидотити, ултрамафични кумулати и амфиболитизирани еклогити. Офиолитите (интензивно разчленени) принципно изграждат фундамента на пъстрите свити.

Метаморфозираните магматити от пъстрите свити формират или послойни тела, алтерниращи с метаседиментите, или като интрузивни тела пресичат ултрамафитите. Главните скалообразуващи минерали на метабазитите са: амфибол + плагиоклаз + кварц + епидот ± гранат ± хлорит. Изчислените температури от 630°C до 520°C при налягания от 6-2 kbar, сочотат умерен афиболитов фациес на метаморфизъм. Установената зоналност на главните скалообразуващи минерали (амфибол, плагиоклаз, гранат) очертава ретрограден ход на метаморфизма.

Химическият състав на изследваните метамагматити ги характеризира като бонинити и островодъгови толеити. За тях са характерни ниски Ti и Zr съдържания, както и типични съотношения на CaO/TiO_2, Al_2O_3/TiO_2, Ti/Zr, Ti/Y, Zr/Y - всичките преходни между островнодъгови толеити и бонинити. Върху редица дискриминационни диаграмии метабазитите от пъстрите свити попадат в полетата на съвременните бонинити и островодъгови толеити. Хондрит-нормираните разпределения на *REE* разкриват присъствието на два тренда: U-образен (типичен за бонинитите) и такъв с недостиг на леки *REE*. Независимо от присъствието на двата тренда, отношението $[La/Sm]_N$ в метабазитите перфектно съвпада с това на много каинозойски бонинитови серии. Метаседиментните скали съдържат теригенни компоненти (метапсамити и кварцити), често алтерниращи с метапелити и мрамори. Естеството на седиментните скални типове отразява техния флишки характер.

Ясните бонинитови и островнодъгово-толеитови особености на магмените скали, както и характерът на седиментните последователности отразяват факта, че пъстрите свити са формирани в условията на островна дъга, надстроена върху океанска кора. Споменатите особености на метамагмените скали свидетелстуват за произхода им в условията на неразвита островна дъга. Характерът на пъстрите свити и тяхната асоциация с блокове от разчленени офиолити показва наличието на сутурна зона. Източнородопската сутурна зона разграничава пъстрите свити от тяхната подложка, изградена от ортогнайси - типични за континентална кора. Съществуващите U-Pb цирконови данни отразяват вариската възраст на ортогнайсите от континеталната кора. Нови U-Pb цирконови определения сочат наличието на късно неопротерозойски протолити сред пъстрите свити.

Introduction

The Variegated Formations of the eastern Rhodope Mountains (SE Bulgaria) form part of the pre-Alpine basement of this region (Kozhoukharov et al., 1992). They are composed of alternating metamorphosed sedimentary and igneous rocks, including metagabbros and amphibolites, felsic orthogneisses, metapelites and metapsammites, and marbles. The total thickness of the Variegated Formations in the area studied is 1800-2900 m, and their rocks record a high-temperature amphibolite facies metamorphic overprint. Numerous ophiolite bodies of the Rhodope ophiolite association (metaperidotites, metacumulates, and amphibolitized eclogites) are associated with the Variegated Formations. The close spatial relationship between the dismembered Rhodope ophiolite bodies and these formations is well known, and the ophiolites form their base (Kozhoukharova, 1996). This poses a question regarding a possible genetic connection between the mafic and ophiolitic rock sequences.

The goal of this paper is to clarify the origin and the significance of the igneous rock assemblages and to determine if there is a connection between them and the adjacent remnants of oceanic crust marked by the Rhodope ophiolite association. The relationship between these rock units has important implications for the geodynamic significance of the structure and evolution of the Rhodope Massif. We have selected rocks from the Variegated Formations of the Avren Synform and the Bela Reka Antiform, close to the Bulgarian-Greek border, to conduct structural, petrographic, and geochemical studies on these assemblages.

The Rhodope massif is part of the Thracian micro continent (after Bončev, 1986), which we regard as a composite superterrane and represents an element of the Variscan belt of Europe. Its pertinence to the mentioned belt is evidenced by features such as the development of voluminous Variscan granitoid magmatism (~340-238 Ma, Zagorčev, Moorbath,

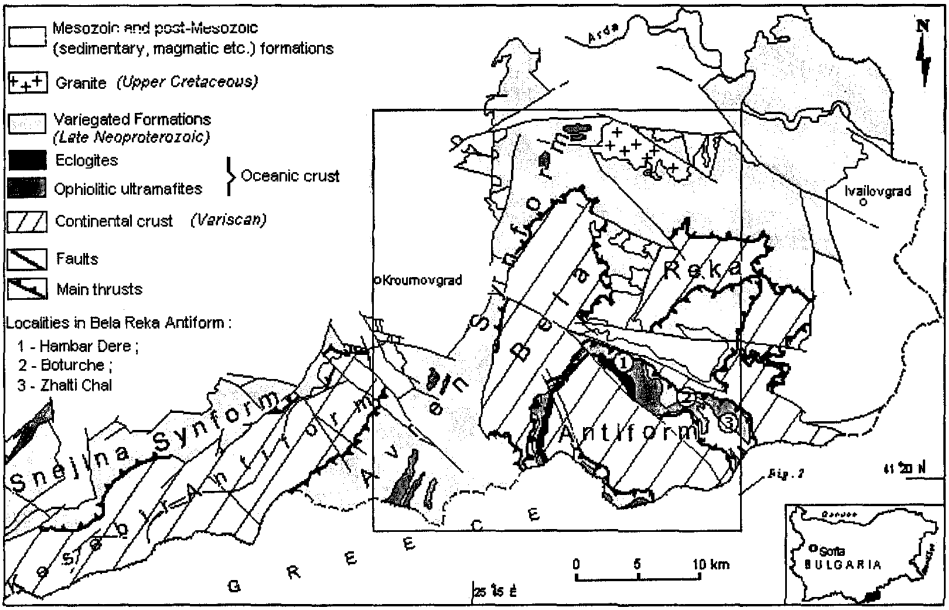

Fig. 1. Geological map of the East Rhodope (after Kozhoukharov et al., 1992) with modifications and additions

Фиг. 1. Геоложка карта на Източните Родопи (по Kozhoukharov et al., 1992) с изменения и допълнения

1986; ~331-250 Ma, Peytcheva, von Quadt, 1995). The Thracian composite superterrane together with the Balkan unit forms the Variscan orogen in Bulgaria.

Geological setting of East Rhodope

The larger bodies of the Rhodope ophiolite association are concentrated in the East Rhodope, as well as the large amounts of metabasic igneous rocks of the Variegated Formation. Additionally, the eastern part of the Rhodope massif has a comparatively lower degree of alpine metamorphism than the central and western parts of the Rhodope massif, where anatexis and migmatization occur in the felsic portions of the sequence. Moreover, the influence of the Late Cretaceous and Tertiary large intrusive bodies in the Rila and Pirin Mountains (West Rhodope) is lacking, and the rocks from East Rhodope are not significantly affected by later thermal perturbations.

Geological features of the Variegated Formations

Several types of sedimentary rocks are recognized among the components of the Variegated Formations. Quartzites and metapsammites alternate with metapelites (Anguelova, Kolcheva, 2001). Marbles, with layers up to 80 m thick, as well as calcschists, are typical for these formations. The wide variety and the composition of these sediments reflect the flysch character of the Variegated Formations (Kozhoukharov, 1987).

The orthoamphibolites occur as layers and slices that are interlayered with the 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). The orthoamphibolites of the Avren Synform have been studied in two localities. The first is the meta-

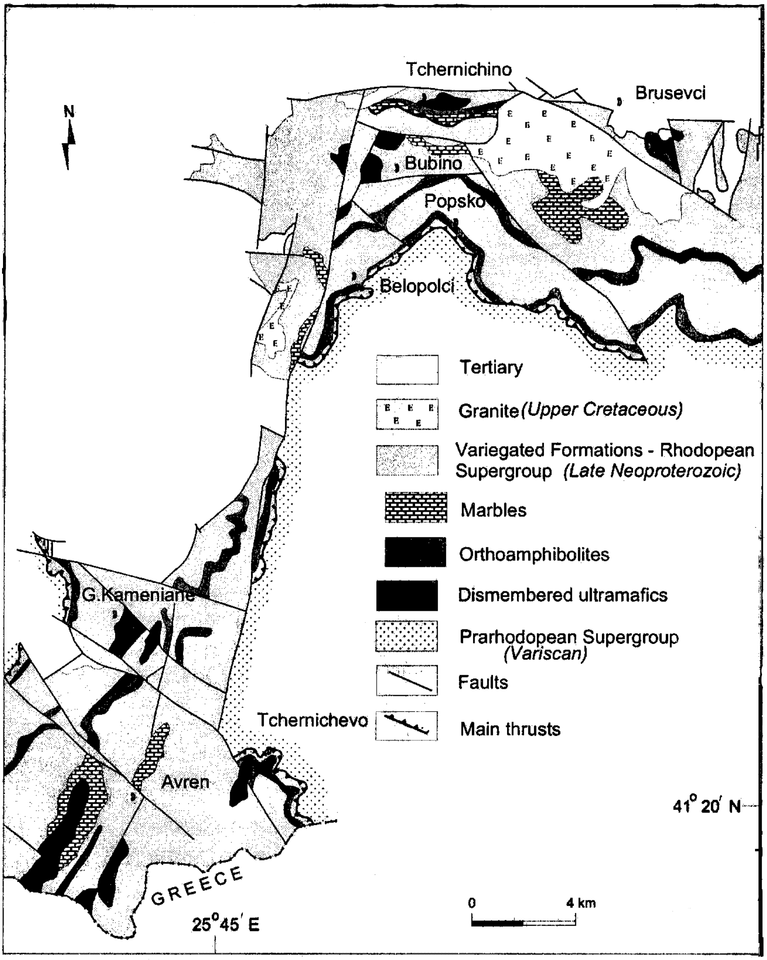

Fig. 2. Geological map of the Avren Synform (after Kozhoukharov et al., 1992) with additions
Фиг. 2. Геоложка карта на Авренската синформа (по Kozhoukharov et al., 1992) с допълнения

gabbro body south of Bubino (Fig. 2), which is transformed into banded amphibolites in its outer parts. This elongated body, with a thickness of ~ 200 m alternates with biotite and two-mica gneisses and with marbles. The second study area is situated directly to the east of the large Avren ultramafic body. In this locality orthoamphibolites alternate with marbles that are part of the marble body shown on Figure 2. Locally the thickness of the ortho-amphibolite layer can attain 100 m.

The relationship between the ortho-amphibolites and the ultramafic rocks was studied in the Bela Reka Antiform, where they build up the limbs of a dome structure (Cara-bunar dome). This relationship was observed in Hambar Dere (1), west of the village of Boturche (2), and to the northeast of the village of Zhalti Chal (3 - see Fig. 1 for map locations). These metabasites intersect the ultramafic bodies, and contain xenoliths of the ultramafic rocks that are typically altered to actinolite-talc or talc-chlorite schists along their edges. In some localities the metamafic rocks from Bela Reka Antiform were metasomatically altered into tourmaline bearing metagabbro-pegmatites, or into clinozoisite-clinopyroxene rodingite-like rocks (Zhalti Chal; Hambar Dere – Fig. 1).

Orthoamphibolites are widespread in the Variegated Formations. They are fine- to medium-grained, mesocratic to melanocratic rocks showing variable amounts of post-magmatic shearing. They are massive in the internal parts of the bodies, and usually foliated in their outer parts, with rare relics of ophitic textures preserved. The igneous activity had multistage character as reflected by the crosscutting relations of compositionally and structurally different dikes and large, irregularly shaped bodies from the Bela Reka Antiform. Fine- to medium-grained melano- to mesocratic metagabbro to metagabbro-dioritic bodies crosscut serpentinized peridotites. The rocks of these bodies are not always homogeneous. We observed melanocratic and leucocratic nebulous portions with rapid transitions between these textures. These bodies are intruded by fine- to medium-grained melanocratic metagabbro or metagabbro-dioritic dikes that also intrude the serpentinites. All rocks of the considered bodies are recrystallized but not intensively foliated. Some of the fine-grained amphibolites, interlayered with parametamorphites, may represent preserved parts of metavolcanic sequence.

Analytical methods

We used JEOL JXA-8800 SuperProbe at the Florida Center for Analytical Electron Microscopy at the Florida International University in Miami to determine the chemical composition of rock forming minerals. Microprobe operating conditions for wavelength dispersive analyses included an accelerating voltage of 15 kV, a 20nA current, and a spot size of 1-2 μm. Counting times were 10s for each element, with a background count of 5 s.

Analyses of whole rock major and trace elements were performed via several methods. Bulk rock major elements were determined by wet chemical analyses, Cu, Zn, Pb, Ni, and Co by AAS using a Perkin-Elmer Spectrophotometer 3030, and Rb, Ba, Sr, Cr, V, Zr, and Y were analyzed by XRF using a VRA-2 spectrometer. All the analyses were performed in the Research Geological Laboratory at the Geological Institute of the Bulgarian Academy of Sciences. Rare earth elements (*REE*) and Y abundances were measured via a HP 4500 plus Series 200 ICP-MS at the College of Marine Sciences, University of South Florida, St. Petersburg, FL, USA. Data were normalized to repeat analyses of the certified geochemical reference samples (USGS basalts: BIR–1, UB-N; NBS688 and W-2), with reproducibility on the order of 5% to 10%.

The rock samples of the investigated orthoamphibolites are deposited in the Geocollections of the Geological Institute (N MER.1. 03. 10)

Petrography and mineral chemistry of the metabasites

The rock forming minerals in the investigated orthoamphibolites are amphibole + plagioclase + quartz + epidote ± garnet ± chlorite. Accessory phases include titanite, apatite, rutile, magnetite, and zircon. Amphiboles are the dominant minerals in most of these rocks. They are sub- to idioblastic green nematoblasts, coarse-grained in Bubino and Avren rocks and medium-grained in the rocks from Bela Reka Antiform. Large S_1-amphibole porphyroblasts (0.2-0.5 cm) occur in Bubino metagabbro. They are surrounded by S_2-prismatic amphiboles, defining clear foliation in the outer parts of the body. The large amphibole porphyroblasts contain numerous rounded quartz inclusions. Very rare garnet grains are included in the periphery of some porphyroblasts. All amphiboles can be classified as Ca-amphiboles according to the classification of Leake et al. (1997) and show a tschermakite to magnesio-hornblende composition (Table 1, Fig. 3a).

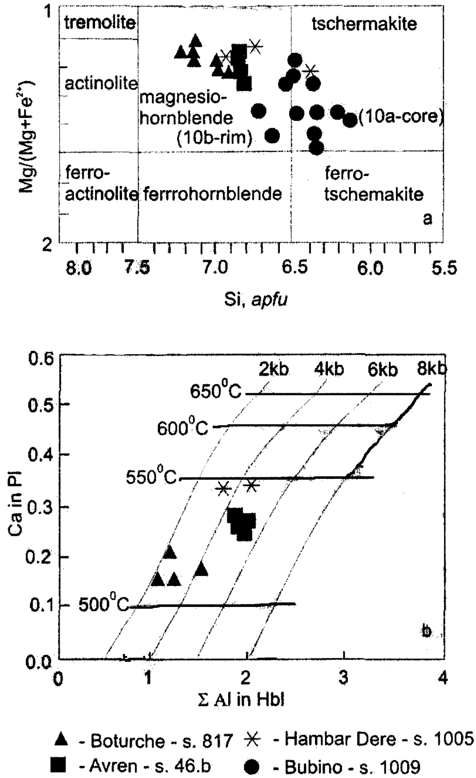

Fig. 3a. Amphibole classification diagram after Leake et al. (1997); b. Graphical geothermobarometer of Plyusnina (1982)

Фиг. 3a. Класификационна диаграма за амфиболите по Leake et al. (1997); b. графичен геотермобарометър по Плюснина (1982)

Amphiboles from Bubino are mainly tschermakites. The fine-grained amphiboles and the rims of some large grains are magnesio-hornblendes. Amphiboles from all other studied localities are Mg-rich hornblende, with exemption of one mineral analysis from Hambar Dere.

Plagioclase crystals from the Boturche and Hambar Dere samples are completely recrystallized into fine-grained aggregates. In contrast, plagioclase grains from the Avren samples are coarse-grained and prismatic. In places from all these localities, magmatic, euhedral grains are partly preserved. An-content ranges from 33.2 to 18.3 mol % (Table 2) for most samples, but high An-contents in the range of 55.9 to 92.3 mol % occur in metagabbros from Bubino. In this body, coarse plagioclase prismatic grains are intensively deformed and recrystallized into subhedral grains with fine polysynthetic deformational lamellae or micro-grained aggregates. The recrystallized outer portions of large plagioclase grains show low An-contents (55.9–66.1 %) while the cores of the grains display higher An-content.

Granoblastic quartz occurs as single grains, or with plagioclase in leucocratic bands in the foliated outer parts of some bodies. Euhedral coarse-grained or anhedral fine-grained epidote and zoisite usually grow at the expense of plagioclase. Garnet occurs very rarely in more leucocratic portions of the metabasic bodies from Hambar Dere and Boturche. In the Bubino metagabbro, garnet is found in the outer, banded parts of the body. Its composition is Alm 61.2–56.3; Gross 20.5–17.4; Prp 20.1–19.2; Spess 3.8–2.1. Pyrope and almandine components decrease slightly from core to rim, while the spessartine component increases slightly.

Metamorphic *P-T* conditions

The absence of relics of igneous minerals and the observed microstructural relations indicate that the protoliths of the studied metamorphic rocks are completely recrystallized and mineral assemblages are re-equilibrated. In some localities, however, traces of magmatic ophitic or porphyritic textures are still preserved, especially in the Bubino gabbro.

The chemical zonation from core to rim in plagioclase, amphibole, and garnet reveals the simultaneous decompression and cooling path of the metamorphic evolution of the investigated rocks. *P-T* estimates are based on mineral phases in equilibrium. Generally, the pressure and temperature determinations of the orthoamphibolites from the localities of Boturche, Hambar Dere, and Avren indicate moderate amphibolite facies metamorphism. The temperatures estimated using the Holland and Blundy (1994) thermometer range from

Table 1. *Chemical composition of selected amphiboles from the metabasic rocks*
Таблица 1. *Химически състав на избрани амфиболи от метабазични скали*

Sample	817-2a	817-2e	1005-1b	1005-2d	46b-2c	46b-3a	1009-11a	1009-10a core	1009-10b rim	1009-3c
Location	Boturche		HambarDere		Avren		Bubino	Bubino	Bubino	Bubino
Nomenclature	Mg-Hbl	Mg-Hbl	Mg-Hbl	Mg-Hbl	Mg-Hbl	Mg-Hbl	Tsch	Tsch	Mg-Hbl	Tsch
SiO_2	50.59	50.94	49.50	47.70	47.50	47.61	44.64	42.87	44.71	43.37
TiO_2	0.15	0.15	0.21	0.28	0.37	0.37	0.21	0.39	0.41	0.41
Al_2O_3	8.15	7.68	10.06	10.85	11.24	11.06	13.90	15.60	11.67	11.83
FeO	12.24	12.08	13.84	14.36	9.71	8.85	14.13	17.29	17.93	17.39
Cr_2O_3	0.15	0.13	0.02	0.01	0.19	0.13	n.d.	0.02	n.d.	0.08
MnO	0.21	0.22	0.29	0.31	0.14	0.17	0.19	0.26	0.23	0.23
MgO	14.33	14.27	13.12	12.74	14.22	14.46	11.04	9.18	9.36	9.95
CaO	9.64	9.45	10.09	9.49	11.89	11.50	10.01	11.41	11.59	11.43
Na_2O	1.63	1.57	1.18	1.61	1.44	1.61	0.92	1.04	0.94	0.89
K_2O	0.22	0.16	0.03	0.11	0.3	0.3	0.27	0.31	0.25	0.22
Total	97.31	96.65	98.34	97.46	97.00	96.00	95.00	98.00	97.09	95.80
TSi	7.134	7.217	6.927	6.743	6.815	6.868	6.477	6.198	6.612	6.463
TAl	0.866	0.783	1.073	1.257	1.185	1.132	1.523	1.802	1.388	1.537
Sum T	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000
CAl	0.487	0.498	0.585	0.549	0.715	0.747	0.852	0.854	0.645	0.539
CCr	0.016	0.014	0.002	0.001	0.022	0.015	0.000	0.002	0.000	0.009
CFe^{3+}	0.934	0.909	1.092	1.312	0.258	0.23	1.203	0.976	0.662	0.951
CTi	0.016	0.016	0.022	0.03	0.039	0.04	0.023	0.043	0.046	0.045
CMg	3.012	3.014	2.737	2.685	3.042	3.11	2.388	1.979	2.064	2.21
CFe^{2+}	0.510	0.522	0.528	0.385	0.907	0.837	0.512	1.114	1.555	1.216
CMn	0.025	0.027	0.034	0.037	0.017	0.021	0.023	0.032	0.028	0.03
Sum C	5.000	5.000	5.000	5.000	5.000	5.000	5.000	5.000	5.000	5.000
BCa	1.456	1.434	1.513	1.437	1.828	1.777	1.556	1.768	1.837	1.825
BNa	0.446	0.431	0.319	0.441	0.172	0.223	0.260	0.232	0.163	0.175
Sum B	1.902	1.866	1.832	1.879	2.000	2.000	1.816	2.000	2.000	2.000
ANa	0.000	0.000	0.000	0.000	0.228	0.228	0.000	0.059	0.106	0.081
AK	0.039	0.03	0.005	0.019	0.055	0.056	0.050	0.057	0.047	0.041
Sum A	0.039	0.03	0.005	0.019	0.284	0.283	0.050	0.116	0.153	0.123
Sum cat.	14.941	14.895	14.837	14.898	15.284	15.283	14.866	15.116	15.153	15.123
X_{Mg}	0.86	0.85	0.84	0.87	0.77	0.79	0.82	0.64	0.57	0.65

Note: n. d.-not detected; structural formulae were calculated on the basis O = 23, 13 CNK, using RECAMP software (Spear, Kimbal, 1984)
Забележка: n. d.-под откриваемия минимум; структурните формули са изчислени на базата на O = 23, 13 CNK, с помощта на софтуер RECAMP (Spear, Kimbal, 1984)

Table 2. *Chemical composition of selected plagioclases from the metabasic rocks*
Таблица 2. *Химични състави на избрани плагиоклази от метабазични скали*

Sample	817-2b	817-2d	1005-1a	1005-4b	46b-2b	46b-3b	1009-4c	1009-10c core	1009-10d rim	1009-3b	1009-1b
Location	Boturche		Hambar Dere		Avren		Bubino				
SiO_2	64.04	63.36	60.04	62.31	61.92	62.01	45.49	47.36	52.91	47.18	53.83
TiO_2	n.d.	n.d.	0.01	0.01	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Al_2O_3	21.64	22.37	24.59	23.03	23.42	22.98	33.80	32.71	29.31	32.39	28.42
FeO	0.11	16.00	6.00	0.14	1.00	0.02	0.15	0.37	0.44	0.21	0.35
MnO	n.d.	0.03	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.05
MgO	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.02	n.d.	n.d.
CaO	3.95	4.59	6.98	5.18	5.65	4.71	18.42	16.49	12.20	16.76	11.69
Na_2O	9.70	9.39	7.75	9.03	8.19	8.56	0.83	1.84	4.44	1.57	5.07
K_2O	0.05	0.06	0.02	0.06	0.14	0.16	0.03	0.03	0.05	0.03	0.03
Total	99.49	99.96	99.45	99.76	99.42	98.44	98.72	98.8	99.37	98.14	99.44
Ab	81.4	78.5	66.7	75.7	71.8	76.0	7.5	16.8	39.6	14.5	43.9
An	18.3	21.2	33.2	24.0	27.4	23.1	92.3	83.0	60.1	85.4	55.9
Or	0.3	0.3	0.1	0.3	0.8	0.9	0.2	0.2	0.3	0.2	0.2

Note: n.d.- not detected
Забележка: n.d. - под откриваемия минимум

630°C to 525°C, and from 550° to 520°C using the Pluysnina (1982) thermometer (Fig. 3b). These results are similar to other authors who have previously reported that the Holland and Blundy (1994) thermometer produces higher temperatures for meta-basites (e.g., Nasir, Okrusch, 1997; John et al., 1999).

Pressure determinations using the Al-in-hornblende barometers and the calibrations of Hammarstrom and Zen (1986), Hollister et al. (1987), and Schmidt (1992) yield mutually comparable pressure values for most samples. For the Boturche locality, the estimated pressures are 4-2 kbar; 5.5-4 kbar for Hambar Dere; and 6-5 kbar for Avren. The pressures after the barometer of Plyusnina (1982) are generally in accordance with the pressure-estimates obtained by the Al-in-hornblende barometers (Fig. 3b).

Pressure and temperature determinations for the Bubino metagabbro are somewhat different. The estimated high pressures (from 11 to 5 kbar), very high An-content in plagioclase (An_{92}–An_{83}) and the high tschermakite substitution in the inner parts of the large amphibole grains show high-grade metamorphic conditions at the amphibolite-granulite facies boundary (see also Bucher, Frey, 1994). The clear foliation in the periphery of the metagabbro body is well marked by S_2 amphibole-garnet-epidote assemblages and plagioclase-quartz bands. Relics of S_1 high An plagioclase are re-oriented along S_2. The temperatures estimated by the Hbl-Pl thermometer of Holland and Blundy (1994) are 780° to 680°C.

Geochemistry

Fifteen orthoamphibolite samples from the Avren Synform and the Bela Reka Antiform were analyzed for major, trace, and rare earth elements. The metamorphism of these rocks obscures their original minerals and textures, and causes problems in classifying them. Nonetheless, evidence for widespread metasomatic alteration is lacking, and the geochemistry of these rocks is expected to mimic

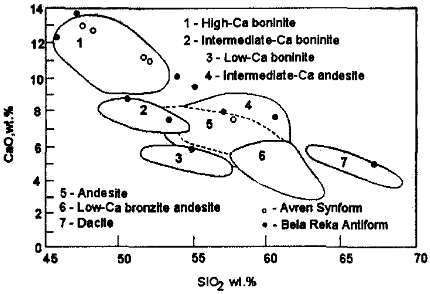

Fig. 4. CaO - SiO_2 diagram for the orthoamphibolite rocks from the Variegated Formations. Fields after Spadea et al. (1998). Symbols are the same in Fig. 5 and 6

Фиг. 4. CaO – SiO_2 диаграма за ортоамфиболитовите скали от пъстрите свити. Полета по Spadea et al. (1998). Същите символи са използвани на фиг. 5 и 6

the original bulk rock chemistry, especially highly immobile elements. Special attention was therefore given to the behavior of the immobile HFSE (high field strength elements) and *REE*, and patterns are compared to similar analogous suites. The most striking features of the rocks studied are their low Ti and Zr contents. Plotted on the TiO_2 vs. Zr discrimination diagram (Pearce, 1980), they fall in the field of volcanic arc basalts (not shown). Characteristics such as CaO/TiO_2, Al_2O_3/TiO_2, Ti/Zr, Ti/Y, and Zr/Y ratios also support an island arc affinity (Table 3). All are transitional between island arc tholeiites and boninites. The samples fall predominantly into the low-Ca boninite group (with $CaO/Al_2O_3<0.75$) according to the classification by Crawford et al. (1989). When plotted in the CaO vs. SiO_2 classification diagram, our samples distribute in the fields for high-Ca boninite, intermediate-Ca boninite, low-Ca boninite, intermediate-Ca andesite, and andesite (Fig. 4, after Spadea et al., 1998). One sample (1004c) falls within the dacite field. The investigated orthoamphibolites have variable MgO contents (from 9.9 to 5.5 wt %), but their Mg-number values (from ~0.75 to ~0.60, except samples 1009d, 1004c and 15b) are close to that of primitive mantle derived magmas (Bloomer, Hawkins, 1987). Elemental characteristics of these samples are similar to Tertiary boninites of Bonin Island, DSDP Site 458, and boninites from Cyprus (Fig. 5a, except s.s. 1004c and 15b). The geochemical features of the orthoamphibolites from Avren Synform and Bela Reka Antiform can be seen on the Mg-number vs. TiO_2 diagram of Pearce et al. (1992), modified by Wyman (1999) with the field of Birch Lake Paleoproterozoic tholeiites (Fig. 5b). Some of the orthoamphi-bolites reside in the fields of DSDP Site 458 and Zambales ophiolite boninites, one - in the Birch Lake tholeiite field. The most primitive samples- 46a, 46b and 75 fall in the MORB

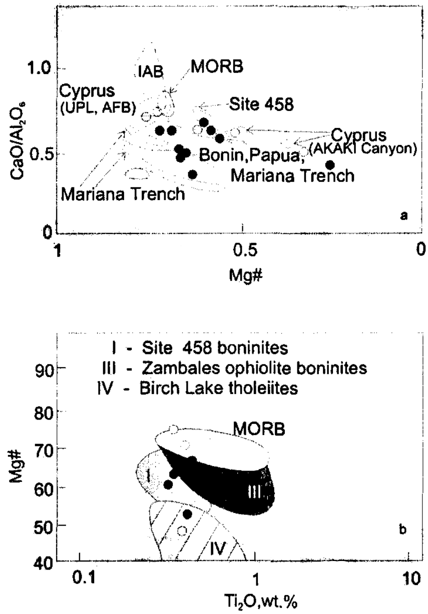

Fig. 5a. CaO/Al_2O_3 – Mg # diagram (fields after Beccaluva, Serri, 1988); b. Mg # - TiO_2 diagram (fields after Pearce et al., 1992, and Wyman, 1999)

Фиг. 5а. CaO/Al_2O_3 – Mg # диаграма (полета по Beccaluva, Serri, 1988); b. Mg # - TiO_2 диаграма (полета по Pearce et al., 1992; и Wyman, 1999)

Table 3. *Chemical composition of metabasic rocks*
Таблица 3. *Химичен състав на метабазични скали*

	Avren Synform					Bela Reka Antiform									
	Avren		Bubino			Hambar Dere				Boturche		Zhalti Chal			
wt. %	46a	46b	15	15b	1009d	1004c	1005c	831	1006	817	817-1	74	75	76a	76b
SiO_2	48.22	47.55	51.63	57.67	52.11	67.30	60.58	57.01	54.86	55.11	53.91	45.73	47.12	53.37	50.50
TiO_2	0.47	0.36	0.01	0.45	0.45	0.68	0.45	0.50	0.36	0.76	0.65	0.48	0.01	0.61	0.40
Al_2O_3	16.55	17.60	17.11	15.31	17.19	11.48	14.01	12.16	15.40	14.35	14.16	20.65	20.91	15.08	17.07
Fe_2O_3	2.20	1.90	3.20	4.90	4.03	6.69	1.81	3.46	1.80	3.56	4.34	3.24	2.34	3.27	3.61
FeO	4,73	4.20	5.47	6.60	6.65	3.36	4.46	4.61	6.92	5.08	4.79	6.16	3.43	4.76	4.74
MnO	0.11	0.11	0.13	0.18	0.19	0.09	0.12	0.13	0.13	0.15	0.19	0.07	0.05	0.12	0.09
MgO	9.07	9.80	7.15	3.05	5.51	1.58	6.21	8.81	7.54	5.94	6.65	5.87	7.14	7.78	7.70
CaO	12.69	12.98	11.12	7.42	10.9	5.04	7.53	7.84	5.65	9.36	9.95	12.34	13.66	7.40	8.58
Na_2O	2.61	2.26	1.36	1.77	1.35	2.76	3.65	3.39	4.39	3.58	3.63	2.18	1.95	4.94	4.44
K_2O	0.30	0.30	0.24	0.33	0.21	0.10	0.10	0.38	0.13	0.16	0.12	0.20	0.13	0.08	0.15
P_2O_5	0.04	0.03	0.03	0.08	0.08	0.11	0.08	n.a.	0.11	0.35	n.a.	0.02	0.02	0.09	0.03
LOI	2.76	2.70	2.43	2.46	1.17	1.40	0.96	0.28	1.96	0.81	1.00	2.82	2.99	2.48	2.50
Total	99.75	99.79	99.88	100.22	99.84	100.59	99.96	98.57	99.25	99.21	99.39	99.76	99.75	99.98	99.81
Mg#	70.71	74.70	60.40	33.00	48.91	23.10	64.49	67.00	61.10	56.10	57.70	53.50	69.70	64.30	63.21
ppm															
Cu	18	54	145	75	72	17	15	27	10	4	5	7	4	6	41
Zn	30	25	59	86	89	159	56	69	232	63	73	15	9	39	39
Pb	7	6	4	4	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	7	8	8	6
Ni	80	120	47	3	15	10	85	143	125	<5	11	14	40	142	80
Co	27	29	40	23	38	<10	26	28	38	31	30	36	26	27	26
Zr	26	16	<7	<7	45	170	100	52	83	185	185	19	17	59	32
Y	12.8*	10.4*	5.9*	15.9*	10	32	42	17	18	28	25	3.3*	6	14.1*	8.8*
Sr	361	273	53	62	120	320	215	140	150	440	370	175	177	115	174
Rb	7	3	27	13	9	3	3	15	3	4	20	3	7	5	3
Ba	86	73	30	79	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	88	56	137	128
Cr	470	557	57	15	13	6	212	373	170	66	75	31	11	256	57
V	172	140	156	200	190	60	130	240	200	210	170	904	259	193	226

Note: n.a.- not analyzed; * - determined by ICP-MS; Mg# =100 × $Mg/(Mg+Fe^{2+})$ - atomic ratios
Забележка: n.a. - не анализиран; * - ICP-MS определение; Mg# =100 × $Mg/(Mg+Fe^{2+})$ - атомни отношения

field. It should be noted, however, that rocks from both the Avren Synform and the Bela Reka Antiform regions fall in the boninitic, tholeiitic and MORB fields of this diagram. Taylor et al. (1992) used Zr/Y ratios to distinguish between arc (Zr/Y< 2) and either fore- or back-arc setting (Zr/Y>2). On this basis the investigated orthoamphibolites from Avren Synform fall in the first group, and these from Bela Reka Antiform in the second one. The rocks from the first group also have higher Ti/Zr and Ti/Y ratios in comparison with those from the second group. And finally, on the TiO_2-10×MnO-10×P_2O_5 discrimination diagram of Mullen (1983), our rocks cluster between the island-arc tholeiite (IAT) and the boninite (BON) fields (Fig. 6).

A characteristic signature of boninite and boninite-like rocks is their chondrite-normalized *REE* patterns. (Table 4). Many authors observed that the U-shaped *REE* pattern is characteristic for the boninites (Hickey, Frey, 1982; Hawkins et al., 1984; Coish, 1989). However, there are also examples of boninitic rocks with light *REE* (*LREE*) depleted patterns (Marianas, New Caledonia, Lau Basin, after Crawford et al., 1989; Cameron, 1989; Hawkins, 1995). The investigated rocks exhibit two distinct patterns (Fig. 7). The first pattern (samples 46a and 46b) shows *LREE* depletions (avg. $[La/Sm]_N$=0.49) and flat to slightly depleted heavy *REE* (*HREE*) patterns (avg. $[La/Yb]_N$=0.44), with distinctive positive Eu anomalies (Eu/Eu* - 1.42 and 1.76 – Fig. 7a). The second group shows characteristic U-shaped boninite-like *REE* patterns (high avg.

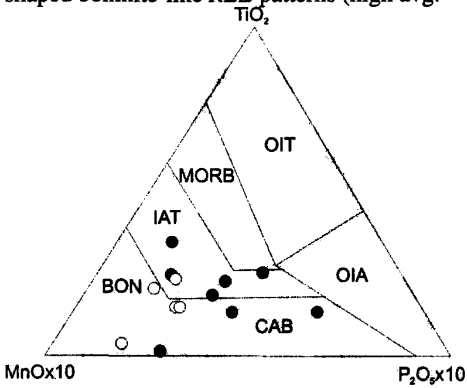

Fig. 6. 10×MnO-TiO_2-10×P_2O_5 discrimination diagram of Mullen (1983). Fields: IAT island-arc tholeiite; BON boninite; CAB calc-alkaline basalt; OIT ocean island tholeiite; OIA ocean island alkali basalt

Фиг. 6. 10×MnO-TiO_2-10×P_2O_3 дискриминационна диаграма по Mullen (1983). Полета: IAT островнодъгови толеити; BON бонинити; CAB калциево-алкални базалти; OIT океански островни толеити; OIA океански островни алкални базалти

Table 3. Notes

Samples: Avren: s. 46a and 46b - mesocratic coarse- to medium-grained metagabbro; Bubino body: s. 15 - porphyritic coarse-grained metagabbro from the central part; s. 15b - partly foliated metagabbro; s. 1009d - foliated and banded metagabbro from the outer part; Hambar Dere: s. 1004c, 1005c and 831 - fine- to medium-grained metagabbrodioritic irregular body, cross-cutting serpentinized peridotite; s. 1006 - melanocratic fine-grained metagabbrodiorite dike, cross-cutting the metagabbrodiorite body; Boturche: s. 817 and 817-1 - melanocratic fine- to medium-grained metagabbrodiorite; Zhalti chal: s. 74 - metagabbroic dike, cross-cutting serpentinized peridotite; s. 75 - metagabbroic irregular body, cross-cutting serpentinized peridotite; s. 76a - fine-grained metagabbroic dike, cross-cutting metagabbroic irregular body (s. 76b), which intersects serpentinized peridotite

Образци: Аврен: обр. 46а, 46b - мезократно грубо до среднозърнесто метагабро; Бубино: обр.15 - порфирно грубозърнесто метагабро от централната част; обр. 15b - частично нашистено метагабро; обр. 1009d - нашистено и ивичесто метагабро от външната част; Хамбар дере: обр. 1004c, 1005c, 831- фино до среднозърнесто метагабродиоритово тяло, секущо серпентинизиран перидотит; обр. 1006 - меланократна финозърнеста метагабродиоритова дайка, секуща метагабродиоритовото тяло; Ботурче: обр. 817, 817-1 - меланократен фин до среднозърнест метагабродиорит; Жълти чал: обр. 74 - метагабродиоритова дайка, секуща серпентинизиран перидотит; обр. 75 - неправилно метагаброво тяло, секущо серпентинизиран перидотит; обр. 76а - финозърнеста метагаброидна дайка, секуща метагаброво неправилно тяло (обр. 76б), което е внедрено в серпентинизиран перидотит

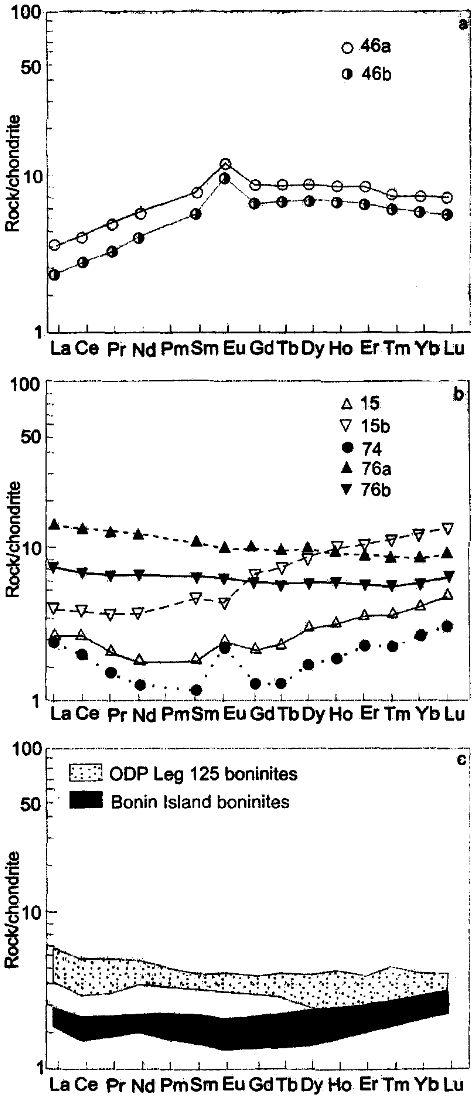

Fig. 7. Chondrite-normalized *REE* pattern for orthoamphibolite rocks studied: a. *LREE* depleted pattern in samples from Avren Synform; b. U-shaped *REE* pattern in samples from Avren Synform and Bela Reka Antiform; c. fields for ODP Leg 125 boninites after Pearce et al. (1992) and for Bonin Island boninites after Shimizu et al. (1992). Data are normalized to C1 chondrite of Sun and McDonough (1989)

$[La/Sm]_N$=1.34), moderately elevated compared to group one but on the whole low (avg. $[La/Yb]_N$=0.87) indicative of both *LREE* and *HREE* enrichment. For this group, varying Eu anomalies from 0.80 to 1.93 were observed (Fig. 7b). The positive Eu anomaly in most of the metabasites suggests plagioclase accumulation. We believe that the analyzed rocks, although metamorphosed to *T* 630-525°C and *P* 6-2 kbar, reflect the *REE* abundances of the protoliths (as shown by Sun, Nesbitt, 1978). Moreover, earlier investigations of Sorensen and Grossman (1989) and Berger et al. (2001 and references therein) indicate that *REE* remain immobile under relatively high-grade metamorphic conditions. On the basis of their *REE* patterns, we believe that samples 15, 15b and 75, which often fall out of the fields of other diagrams, are in fact boninites. Despite the differences in the *REE* patterns in the studied rocks, the comparison with the Tertiary West Pacific boninites (Fig. 7c) shows that our rocks overlap the fields of ODP Leg 125 (Pearce et al., 1992) and Bonin Island boninites (Shimizu et al., 1992). Similar diverse REE patterns were reported for Nepoui and Koh boninite sequences from New Caledonia (Cameron, 1989).

Regardless of some minor differences in the chemical composition of the rocks from the Avren Synform and the Bela Reka Antiform, they both preserve evidence for protoliths with island arc tholeiite and boninite character. According to Bazylev et al. (1999), parts of the ultramafic bodies and metatholeiites were formed in a supra-subduction zone setting.

Фиг. 7. Хондрит-нормирано разпределение на *REE* за изследваните ортоамфиболитови скали; a. обеднено на леки *REE* разпределение в проби от Авренската синформа; b. U –образно разпределение на *REE* в проби от Авренската синформа и Белоречката антиформа; c. поле на ODP Leg 125 бонинити по Pearce et al. (1992) и на бонинити от остров Boinin по Shimizu et al. (1992). Данните са нормирани към C1 хондрита от Sun & McDonough (1989)

Table 4. *REE content in the metabasic rocks, ppm*
Таблица 4. *Съдържание на REE в метабазичните скали, ppm*

	Avren Synform				Bela Reka Antiform		
	46a	46b	15	15b	74	76a	76b
La	0.88	0.55	0.64	0.94	0.62	3.11	1.66
Ce	2.38	1.75	1.60	2.35	1.34	7.55	4.02
Pr	0.46	0.31	0.20	0.34	0.15	1.12	0.60
Nd	2.68	1.92	0.85	1.73	0.64	5.35	2.95
Sm	1.12	0.85	0.28	0.72	0.18	1.60	0.93
Eu	0.63	0.61	0.14	0.25	0.14	0.54	0.36
Gd	1.69	1.34	0.45	1.32	0.27	2.01	1.20
Tb	0.31	0.25	0.09	0.27	0.05	0.34	0.21
Dy	2.13	1.73	0.74	2.15	0.45	2.26	1.44
Ho	0.45	0.37	0.18	0.52	0.11	0.48	0.31
Er	1.31	1.10	0.60	1.75	0.39	1.46	0.95
Tm	0.18	0.15	0.09	0.28	0.06	0.21	0.14
Yb	1.18	0.98	0.72	2.02	0.48	1.45	0.97
Lu	0.18	0.14	0.12	0.33	0.08	0.22	0.15

Discussion

The comparison of the investigated rock assemblages with sequences characteristic of certain geodynamic settings (i.e. island-arc, forearc, back-arc, mid-ocean ridge) is an important tool for identification of their genetic identities.

We propose an origin for the Variegated Formations and part of the associated Rhodope ophiolite association as an ensimatic island arc model (Haydoutov et al., 2000; 2001). Boninite generation is observed during the initial stages of the subduction (Crawford et al., 1989; Pearce et al., 1992). Our model fits well with the widely accepted early-arc development and the associated rock types found in the Izu-Bonin-Mariana subduction system (Bloomer et al., 1995). Some facts indicate the presence of different types of oceanic crust that are identified as Rhodope ophiolite association. The island-arc igneous boninites and tholeiites do not intersect the eclogites, and some of the ultramafic rocks of the Rhodope ophiolite association have been proposed to also form in a supra-subduction zone setting (Bazylev et al., 1999). Alternatively, eclogites from the Rhodope massif have affinities similar to typical MORB (Kolcheva, Escenazi, 1988). The eclogite/ophiolite/boninite ensemble could be a tectonic association of two types of ocean crust, the first one formed at a mid-ocean ridge and underwent eclogite facies metamorphism, and the second formed in a supra-subduction zone setting and underwent later (Variscan?) amphibolite facies metamorphism. A scenario of repeated subduction episodes or subduction polarity reversal could accommodate our model.

The characterization of the Avren Synform sediments as metamorphosed flysch containing a greywacke level (Kozhoukharov, 1987) is an important feature, because such sequences are characteristic of island-arc environments. Examples of such flysch successions are exposed on the Mentawai Islands and Barbados Island (Mitchell, Reading, 1971). Limestones, in association with flysch, are also typical of island arc related successions, especially those in proximity to the island arcs. One example is the Eocene-Pleistocene sedimentary-volcanic sequence formed in the collision zone between the North d'Entrecasteaux Ridge and the New Hebrides (Vanuatu) Island Arc (ODP Site 829, Reid et al., 1994). The association of sedimentary rocks with boninitic and tholeiitic magmatites is typical for the successions of island-arc settings - for example, in the Bonin trench (Taylor et al., 1994). They are also found in ancient island arcs, e.g. those connected with the ophiolites Koh (Meffre et al., 1996) and Bets Cove (Coish, 1989), the Cambrian island-arc in Tasmania (Brown, Jenner, 1989), as well as the Early Proterozoic Trans-Hudson orogen (Wyman, 1999).

The problem for the age of the Variegated Formations is important but in the same time protolith ages for these rocks are almost totally unknown. The new U-Pb zircon dating suggests that the orthoamphibolites of these formations crystallized in the Late Neoproterozoic, 572±5 Ma (Carrigan et al., 2003), but this remains to be proved with more determinations. Obviously the young ages established in the Rhodope metamorphics (e.g. Liati, Gebauer, 1999; Wawrenitz, Mposkos, 1997) are result of the Alpine metamorphic overprint.

The eastern part of the Rhodope Massif consists of antiformal cores (Bela Reka and Kesebir) built up by metagranites, orthogneisses and gneiss-schists (Kozhoukharova et al., 1988; Macheva, Kolcheva, 1992) that are typical of continental crust (Fig. 1). This continental crust forms the Prarhodopian (pre-Rhodope) Supergroup of Kozhoukharov et al (1992). Existing U-Pb zircon data demonstrate that the gneissic protoliths are Variscan in age (~305-320 Ma, Peytcheva, von Quadt, 1995). The synforms (Avren and Snejina) are built up by the double-layered structure. The lower layer consists of fragments of oceanic crust (intensely dismembered, Kolcheva et al., 2000) overlain by the orthoamphibolites and the sedimentary components of the Variegated Formations.

The whole double-layered assemblage is thrusted over the continental crust (Fig. 1). The contact between these two types of crust is tectonic, and the double-layered assemblage is allochthonous. In some localities, ultramafics and eclogites are in direct tectonic contact with orthogneisses, and shearing in the orthogneisses becomes mylonitic as the contact is approached. The Variegated Formations and the Rhodope ophiolite association do not show any evidence of contact metamorphism. The rocks of the continental crust however are intensively sheared. In most of the observed localities the contact bears features of a deep tectonic structure for which internal tectonic imbrication is typical. A zone of intensively sheared granitoids from 100 to 300 m thick exists along the contacts. Rock slices from both types of crust in the zone of tectonic imbrication form a layer thick from 100 to 400 m. The position of the allochthon delineates the East Rhodope suture. Along this suture, the oceanic crust of probable Neoproterozoic age is emplaced over the continental crust of Variscan age. The surface of the suture zone is an intensely folded, sub-horizontal plane in the East Rhodope block. The suture was formed probably in Late Variscan time or later (?).

The ophiolite fragments from the Rhodope massif have been considered as obducted ocean crust (Kozhoukharova, 1985; Kolcheva, Eskenazi, 1988). Prior to obduction, the two types of oceanic crust were tectonically associated. Ricou et al. (1998) distinguished continental and mixed units within the Rhodope massif, the latter containing ophiolites. The close genetic and spatial relations of the ophiolites and the Variegated Formations are a reason to consider these formations as element of the same suture. This is an important feature of the suture taking into account the highly dismembered character of the oceanic crust that underlies the Variegated Formations. The configuration of the suture, mentioned by Burg et al. (1996) for the East Rhodope, coincides with our understanding of this structure. However, Burg et al. (1996) describe an upper terrane with "mafic-ultramafic-gneiss sequence" as well as intermediate trust sheets with several sequences including "eclogite-metabasic-gneiss sequence", and they do not distinguish between the suture and their "syn-metamorphic nappe complex".

Conclusions

The boninitic and island-arc tholeiitic characteristics of the igneous rocks, the flysch properties of the sedimentary components, the presence of considerable amount of carbonates, and finally the association with ultramafic rocks, all suggests that the Variegated Formations were formed as an ensimatic island arc. The described cross-cutting relation of the orthoamphibolites with the ultramafic rocks of Rhodope ophiolite association and the formation of this ultramafics in a supra-subduction zone setting, all indicate possible genetic connection between the Variegated Formations and the mentioned relics of the oceanic crust.

The East Rhodopean suture zone separates the Rhodope terrane, built up by the Rhodope ophiolite association and the Variegated Formations (Rhodopian Supergroup of Kozhoukharov et al., 1992) from the Bela

Reka terrane formed by the Pre-Rhodope (Prarhodopian, after Kozhoukharov et al., 1992) Supergroup. We therefore consider the Rhodope massif as a composite terrane, and the Thracian micro continent of Bončev (1986) as a composite superterrane.

We suggest that the genetic unity of the part of the ophiolites and the Variegated Formations clarify the origin and structure of the Rhodope Massif. Based on regional correlations, the concept of a suture zone could be useful for clarifying the structure of the whole Rhodope composite terrane. The data presented in Daieva and Pristavova (1998) regarding the existence of arc tholeiites and boninites from the Central Rhodope is an important clue for the correlation of the Variegated Formations from both regions and as a consequence for clarifying their structure and evolution.

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