ГОДИШНИК НА МИННО-ГЕОЛОЖКИЯ УНИВЕРСИТЕТ "СВ. ИВАН РИЛСКИ", Том 49, Св. I, Геология и геофизика, 2006
ANNUAL OF THE UNIVERSITY OF MINING AND GEOLOGY "ST. IVAN RILSKI", Vol. 49, Part I, Geology and Geophysics, 2006

GEOCHEMICAL AND SR-ND ISOTOPE CONSTRAINTS ON THE LATE CRETACEOUS MAGMATISM IN THE AREA OF THE ZIDAROVO ORE FIELD

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ABSTRACT. The Zidarovo ore field is situated 15–20 km south from the town of Bourgas. The region consists of Senonian volcanic, intrusive and sedimentary rocks. The syn-/postmagmatic hydrothermal activity was responsible for the formation of polymetallic and gold-polymetallic ore veins in the volcanic rocks. The ore mineralisation is developed in two sectors – Kanarata in the central part (close to the Zidarovo intrusive) and Yurta in the NNW part of the ore field. The composition of the volcanic rocks of the Zidarovo ore field varies from basaltic to trachy-andesitic. The sub-volcanic dyke complex has basaltic and trachy-basaltic composition, and the intrusive rocks are gabbroic to monzo-dioritic. The rock varieties belong to the high-K calc-alkaline magmatic series. Indicative bivariate diagrams as Nb vs. Y, Rb vs. (Y+Nb), Rb vs. (Yb+Ta) and Zr/Al_2O_3 vs. TiO_2/Al_2O_3 define volcanic-arc tectonic environment of their formation. MORB normalized trace elements patterns of the magmatic rocks (enrichment of large ion lithophile elements and low values for high field strength elements, the strong negative Ta and Nb anomaly and the chondrite-normalized REE distribution of the basaltic rocks indicate subduction related magma affinity. Nd and Sr whole rock isotope data are in agreement with this conclusion revealing mantle-crustal signatures with ε-Nd (80 Ma) values generally between +2.1 and +3.0 and initial strontium ratios in the narrow range 0.704-0.705. Compared with the data for the Central Srednogorie the magmatic rocks from the Zidarovo ore field are less crustal contaminated. The same trend is observed comparing intrusive and volcanic varieties - the volcanic rocks reveal more primitive isotope composition, which is explained by the faster cooling of the volcanites, probably prior to contamination with crustal materials in middle/upper crustal chambers.

ГЕОХИМИЧНИ И SR-ND ИЗОТОПНИ ХАРАКТЕРИСТИКИ НА ГОРНО КРЕДНИЯ МАГМАТИЗЪМ В ЗИДАРОВСКОТО РУДНО ПОЛЕ

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РЕЗЮМЕ. Зидаровското рудно поле е разположено на около 15–20 км. южно от гр. Бургас. Районът е изграден предимно от сенонски вулканогенни, интрузивни и седиментни скали. Резултат от постмагматични хидротермални процеси са жилите с полиметални и злато-полиметални руди във вулканските скали. Рудната минерализация е развита в два участъка – Канарата в централната част (в близост до Зидаровския интрузив) и Юрта в север-северозададаната част на руднотото поле. Съставът на вулканските скали в Зидаровското рудно поле варира от базалти до трахиандезити. Субвулканският дайков комплекс има базалтов и трахибазалтов, а интрузивните скали - габров до монцо-диоритов състав. Скалите принадлежат основно към високо калиевата калциево-алкална магматична серия. Индикативни диаграми като Nb vs. Y, Rb vs. (Y+Nb), Rb vs. (Yb+Ta) и Zr/Al_2O_3 vs. TiO_2/Al_2O_3 определят островно-дъгова обстановка на формиране. MORB нормализираните разпределения на редките елементи в скалите (обогатяване на LILE и ниски стойности за HFSE), добре изразената негативна Ta и Nb аномалия и хондрит-нормализираните разпределения РЗЕ (обогатяване на LREE) определят субдукционна обстановка на формиране. Nd и Sr изотопни данни подкрепят този извод като показват мантийно-корови характеристики с ε-Nd (80 Ma) стойности основно между +2.1 и +3.0 и начални стронциеви отношения предимно в тесния интервал 0.704-0.705. В сравнение с данните от централното Средногорие магматичните скали от Зидаровското рудно поле са по-малко корово контаминирани. Същият тренд се наблюдава при сравняването на интрузивните и вулкански скали – вулканските скали показват по-примитивни изотопни състави, което се обяснява с по-бързата кристализация на вулканитите, вероятно преди контаминирането им с корови материали в средно/горно корови магмени камери.

Introduction

Europe's world-class copper-porphyry and Au-epithermal deposits are hosted by an elongated belt of intensive Late Cretaceous magmatic activity, known as the "Carpathian-Balkan Segment" of the "Tethyan Eurasian Metallogenic Belt" (JANKOVIC, 1976 and 1977), the "Banatitic Magmatic and Metallogenic Belt" (e.g. BERZA *et al.,* 1998, CIOBANU *et al.,* 2002) or the "Apuseni – Banat – Timok - Srednogorie belt" (ABTSB) (POPOV *et al.,* 2000, 2003). An abundant new data on the geodynamic control of various ore deposits, the geochronology and geochemistry of the Late Cretaceous magmatism and the specific features of the related Cu-Au deposits in ABTSB have been added during the activity of the GEODE (GEodynamic and Ore Deposit Evolution) project funded by the European Science Foundation. In the

Srednogorie zone they were concentrated in the central part of the zone – the Panagyurishte region (see summary of VON QUADT *et al.,* 2005 and references there), as it hosts the most important economic deposits. Nevertheless the most extensive magmatic activity is manifested in the Eastern Srednogorie zone. The study of the latter helps to define the sources of the magma and allows understanding the geodynamic evolution of the region in Upper Cretaceous time.

In the present study we focus on the petrological and isotope-geochemical characteristics of the Zidarovo ore field (ZOF), as it hosts economic mineralization and provides good opportunity for studying of the link between the geodynamic settings, magmatism and ore formation. The aim of our investigation is to reconstruct the geological evolution of the Late Cretaceous magmatic complex, to identify the temporal and genetic relationships between its magmatic products and the mineralized zones of the ZOF and at least to define the tectonic evolution of the Late Cretaceous magmatic rocks in the area. We have combined field observations with representative whole rock major and trace element analyses and isotope Sm-Nd and Rb-Sr studies to define the magmatic sources and reconstruct the processes that produced the deposit.

Geological background and sampling

The Eastern Srednogorie zone is characterised by the most significant presence of volcano-intrusive structures in the Srednogorie zone. Intensive volcano-tectonic faulting, most often of radial-concentric type provides a suitable condition for precipitation of vein type ore bodies. This part of the Srednogorie zone hosts several ore districts and numerous ore deposits and occurrences. In the last 50 years they were the base for copper production as well as gold, silver, molybdenum and other elements as by-products.

The Bourgas ore region is located in the easternmost part of the Srednogorie zone. The copper ore deposits in this region are genetically related to the Late Cretaceous volcano-plutonic magmatism. The ZOF is situated 15 – 20 km south from the town of Bourgas in the area of Zidarovo, Izvor, Dimchevo and Krushevec villages. Its formation is determined by the development of the central Zidarovo volcano-plutonic structure. The latter is initially mentioned by STANISHEVA-VASSILEVA AND VASSILEV (1972) and is later characterized by RASHKOV *et al.* (1978) and POPOV (1981). The syn-postmagmatic hydrothermal activity was responsible for the formation of polymetallic and gold-polymetallic ore veins in the volcanic rocks. The ore mineralsation is developed in two sectors – Kanarata is in the central part (close to the Zidarovo intrusive) and Yurta is in the N-NW part of the ore field. These two sectors have a different mineral composition – Cu-Bi ore types predominate in Kanarata, whereas Cu-polymetallic mineral assemblages with gold are typical for Yurta (POPOV *et al.,* 1993).

The region consists of Senonian volcanic, intrusive and sedimentary rocks (Fig. 1). ANTONOV *et al. (*1979) and POPOV *et al.* (1980) subdivided them into an olistostrome unit and Kasaldjishka, Novopanicherevska and Rosen formations. According to POPOV (1980) three stages of the Late Cretaceous magmatic activity are distinguished: effusive, sub-volcanic and intrusive. These stages completely cover the

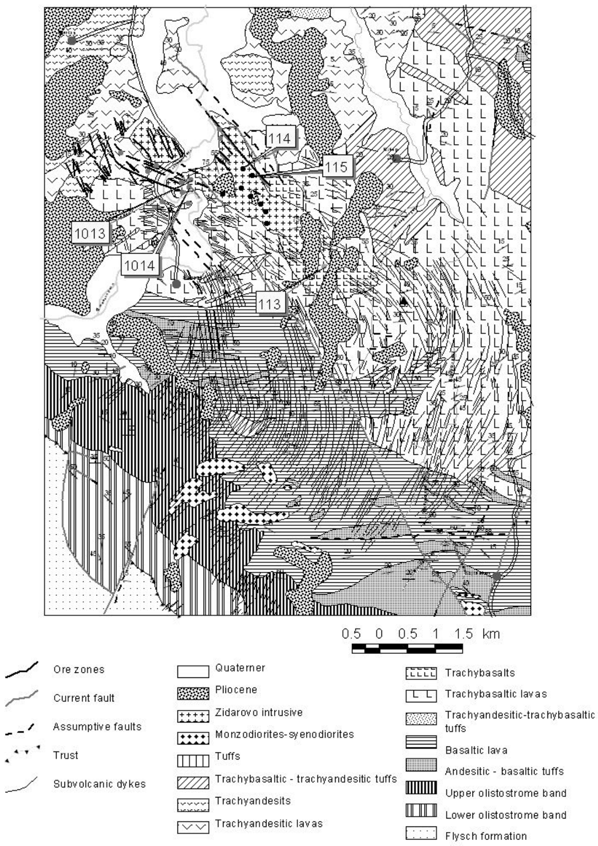

Fig. 1. Geological map of the Zidarovo ore field (after RASHKOV *et. al.,* 1978) with the location of some representative samples

description of the trachyandesitic-trachybasaltic, basaltic-trachybasaltic and gabbro-syenitic stages of RASHKOV *et al.* (1978). During these consecutive stages the Zidarovo volcano, Zidarovo dyke ring complex and Zidarovo intrusive formed, as parts of the complex Zidarovo volcano-plutonic structure (POPOV, 1981). The volcanic rocks here are developed as a 2000–2500 m thick complex of intercalated lava flows and pyroclastic materials (POPOV, 1981). The lava flows have a concentric orientation and dominate in the lower and intermediate parts of the section. A neck and subvolcanic bodies mark the position of the main magma channel. The rocks of Zidarovo volcano could be correlated with the Vurly briag volcanites to the north and the Rosen volcanites to NE and E (POPOV *et al.,* 1983). The volcanogenic sequence consists of trachy-andesites, trachybasalts, latites and trachytes (POPOV *et al.,* 1983), and the composition of the dykes is analogous to the volcanic rocks. The Zidarovo intrusion penetrates the rocks of the effusive and dyke complex. It is emplaced in the central part of the caldera structure (Fig. 1) as an elongated to the north-west body (MARINOV AND BAJRAKTAROV, 1981, POPOV, 1981) and according to MARINOV (1980) consists of monzonites, monzodiorites, essexites, and alkaline quartz syenites.

Analytical techniques

The whole-rock major elements were analyzed by X-ray fluorescence (XRF) in the University of Salzburg, Austria. The trace and rare earth elements (REE) were analyzed by Laser ablation-inductively coupled mass spectrometry (LA-ICPMS) in the laboratories of the Institute of Isotope Geology and Mineral Resources, ETH Zurich, Switzerland. Sm-Nd and Rb-Sr data are measured with the isotope dilution technique.

Chemical composition of the rocks from Zidarovo ore field

The composition of the volcanic rocks and the dykes in the ZOF varies from basaltic to trachyandesitic and trachytic (Table 1, Fig. 2). On the SiO_2-K_2O diagram studied samples are classified as high-K calc-alkaline rocks (Fig. 3). SiO_2 content vary from 47.8 to 60 wt. % and show a positive correlation with the alkali content (Fig. 3).

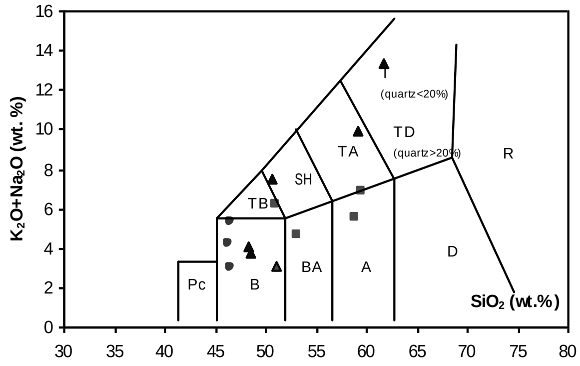

Fig. 2. TAS diagram after LE MAITRE (1989) for representative samples from the ZOF: volcanites (triangles), subvolcanic rocks (circles), intrusive rocks (squares). B – basalt; BA – basaltic andesite; A – andesite; D – dacite; SH – shoshonite; TA – trachy-andesite; TD – trachy-dacite; T – trachyte; Pc - picro-basalt; TB – trachy-basalt; R – rhyolite

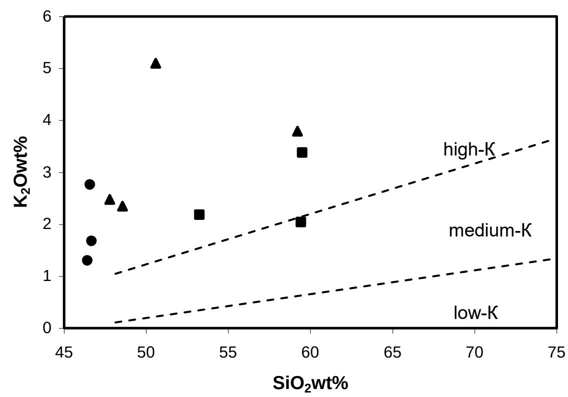

Fig. 3. SiO_2 vs. K_2O diagram (LE MAITRE (1989) for representative samples from the ZOF. Symbols as in Fig. 2

Table 1

Major element composition of the representative samples

Oxides wt. %	1013 volcanic	1014b volcanic	113 subvolcanic	114 intrusive	115 intrusive
SiO_2	60,07	51,11	50,49	59,01	50,98
Ti_2O	0,43	0,56	0,75	0,55	0,68
Al_2O_3	16,88	11,30	14,44	14,63	17,73
Fe_2O_3	5,61	11,47	10,35	7,05	6,43
MnO	0,02	0,19	0,11	0,1	0,08
MgO	0,23	8,84	5,94	3,78	4,3
CaO	0,08	10,06	9,21	6,96	8,04
Na_2O	0,29	2,76	2,51	2,78	5,32
K_2O	13,43	0,36	2,77	2,78	0,88
P_2O_5	0,23	0,33	0,68	0,52	0,75
LoI	2,67	3,04	2,17	1,89	3,99
Total	99,94	100,02	99,42	100,05	99,18

Table 2

Trace element composition (in ppm) of the representative samples

Elements	1013	1014b	113	114	115
Sc	9.6	42.3	37.0	33.0	25.5
V	156.6	268.5	324.7	349.9	309.9
Cr	49.6	392.6	92.3	59.0	61.7
MnO	0.1	0.2	0.1	0.1	0.1
Co	2.4	42.8	35.4	30.3	17.1
Ni	19.5	87.4	24.8	20.0	15.5
Cu	577.8	36.1	90.4	286.3	4.6
Zn	111.5	60.5	39.3	68.9	57.4
Ga	n. d.	n. d.	12.9	17.4	15.9
As	4.6	12.1	241.8	318.7	194.6
Rb	147.7	8.7	55.9	99.0	17.1
Sr	628.2	812.1	968.3	970.1	949.7
Y	11.4	14.3	18.5	21.5	21.7
Zr	74.2	37.5	48.2	54.7	73.7
Nb	3.6	1.5	2.1	2.4	3.0
Mo	1.5	0.6	0.6	1.3	0.7
Cs	1.1	0.2	2.0	4.4	1.6
Ba	1158.1	47.5	310.5	465.1	133.4
La	16.8	10.8	12.5	16.3	14.4
Ce	35.2	22.8	25.5	34.6	28.6
Pr	4.0	3.0	3.6	4.5	3.5
Nd	17.4	15.6	17.0	19.7	16.2
Sm	3.2	3.6	4.1	4.6	4.2
Eu	0.9	1.4	1.4	1.3	1.3
Gd	2.0	3.6	4.2	4.1	4.0
Tb	0.3	0.5	0.6	0.6	0.6
Dy	2.1	3.0	3.4	3.8	3.7
Ho	0.4	0.5	0.7	0.8	0.7
Er	1.0	1.5	1.8	2.2	2.1
Tm	0.2	0.1	0.2	0.3	0.3
Yb	1.2	1.2	1.8	2.3	2.4
Lu	0.2	0.1	0.3	0.3	0.3
Hf	1.6	1.0	1.5	1.7	2.1
Ta	0.2	0.1	0.1	0.2	0.2
W	2.0	0.5	0.9	3.4	1.7
Pb	124.2	10.1	44.4	51.2	34.3
Th	5.2	1.8	3.0	8.3	8.3
U	4.9	0.6	1.2	2.8	3.7

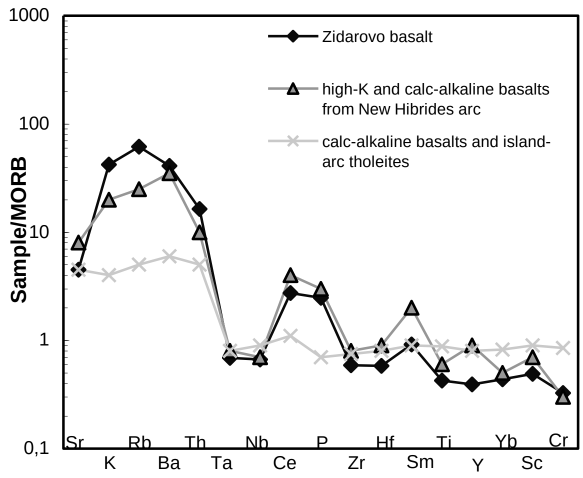

Fig. 4. MORB-normalized spider diagram (after PEARCE, 1983) for the investigated basalts from the Zidarovo deposit compared with typical basalt series related to subduction settings. High K calc-alkaline basalts of the New Hibrides arc from KAMENOV, 2003

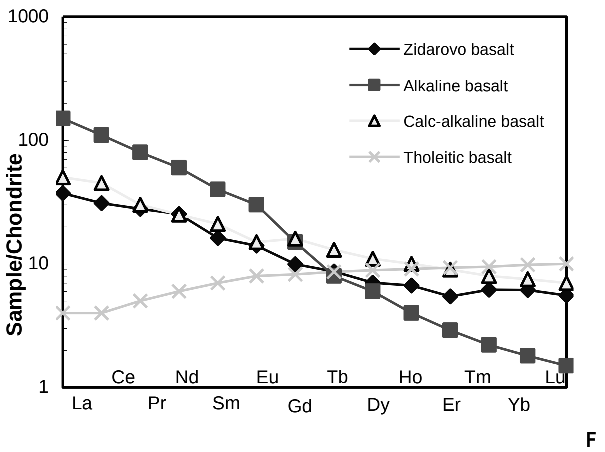

Fig. 5. Chondrite normalized REE patterns of the basalts from Zidarovo ore field compared with additional different basaltic rocks (values from KAMENOV, 2003). Normalizing values after NAKAMURA, 1974

MORB normalized patterns for the magmatic rocks (Table 2, Fig. 4) indicate an enrichment of large ion lithophile elements (LILE) and low values for HFSE (high field strength elements – Ce, P, Zr and Hf). The strong negative Ta and Nb anomaly indicate subduction related magma affinity (Fig. 4). The elements with lower ionic potential (K, Na, Rb, Ba and Sr) show considerable variations. Ba values vary between 133 and 465 ppm, Rb is from 17 to 99 ppm and Sr from 892 to 1176 ppm in the Zidarovo intrusive rocks. In the dyke rocks these values are respectively between 297 and 310, 56-65, and 477-968, whereas for the volcanic rocks these intervals are 47-1504, 8-194, and 103-1060.

The LREE enrichment ranges from 20 to 51 (Fig. 5), whereas La_n/Yb_n ratios vary from 4 to 8. Middle and heavy REEs show relatively flat patterns, which are generally within 4–10 times that of chondrite. The dyke rocks show lower values of LREE compared with the intrusive rocks from the Zidarovo. The lack of Eu anomaly suggests that there was no plagioclase fractionation involved during the genesis of the volcanic rocks.

Comparing MORB-normalized trace element distribution of basalts from ZOF with typical basalt series in subduction settings a similarity with high-K and calc-alkaline basalts is observed (Fig. 4). The chondrite normalized REE pattern of the basalts from ZOF resembles this of calc-alkaline basalts (Fig. 5).

The Zr/Al_2O_3-TiO_2/Al_2O_3 diagram for potassic volcanic rocks from different tectonic settings (MŰLLER *et al.,* 1992) is used to discriminate within-plate rocks from oceanic arc rocks. The studied volcanic rocks from the ZOF fall in the fields of continental and postcollisional arcs (Fig. 6a). At the Ce/P_2O_3-Zr/TiO_2 discrimination diagram most of the samples falls in the field of the poscollisional arcs (Fig. 6b).

For distinguishing of granites from known tectonic settings the elements Rb, Y (and its analogue Yb) and Nb (and its analogue Ta) were selected as the most efficient discriminates. Discrimination diagrams of Pearce et al. (1984) are used, which are primary supposed to classify granites into ocean-ridge, volcanic-arc, within-plate and collisional types. A bivariate plot of Nb and Y is subdivided into three fields (Fig. 6c) – oceanic-ridge granites (ORG), within-plate granites (WPG) and volcanic-arc granites (VAG) together with syn-collisional granites (syn-COLG). A plot of Ta and Yb is similar and allows the fields of syn-collisional and volcanic-arc granites to be separated (Fig. 6d). The discrimination diagram Rb vs. (Y+Nb) and Rb vs. (Yb+Ta) more efficiently separate syn-COLG from VAG and also gives clear division between WPG and ORG (Fig. 6e, f). Supra-subduction zone granites can fall in the VAG field. They may be identified on an Nb-Y diagram as they usually reveal lower Y content (Rollinson, 1993). On all diagrams the samples from Zidarovo magmatic rocks belong to the field of VAG.

Sr-Nd isotope data

Sm-Nd and Rb-Sr isotopes are measured on samples from ZOF which are characterized by detailed petrological studies. Preliminary $^{143}Nd/^{144}Nd$ data for the intrusive rocks give evidence for mantle dominated magma source and the ε-Hf (corrected for 80 Ma) values range between 2.14 and 3.00. Similar values of ε-Nd in the gabbro and monzodiorite argue for a common source for both rock varieties. Compared with the data for the Central Srednogorie (VON QUADT *et al.,* 2005) the rocks of Eastern Srednogorie crystallize from more primitive mantle magmas and are less contaminated with continental crust materials. These conclusions are confirmed by the Sr isotope data. The initial strontium ratio in the Late Cretaceous rocks of ZOF lay in a narrow range of 0.704-0.705 and only two out of ten samples reveal $(^{87}Sr/^{86}Sr)_i$ close to 0.706. The dykes and volcanics are less enriched in radiogenic Sr, which emphasize the upper crust as most probable source of contamination in the intrusive rocks. The volcanic rocks on the other hand are less contaminated, as they are cooled faster and probably prior to contamination with crustal materials.

Conclusions

The magmatic rocks of Zidarovo ore field show wide compositional variation with some differences between the volcanic, subvolcanic and intrusive rocks. The major element compositions of the volcanic rocks define them as basalts to trachyandesites, the subvolcanic dyke complex has basaltic and trachybasaltic composition, and the intrusives are gabbros to diorites. All rock varieties belong to the calc-alkaline and high-K calc-alkaline magmatic series. Despite some variations in trace and REE-elements compositions Indicative bivariate diagrams as Nb vs. Y, Rb vs. (Y+Nb), Rb vs. (Yb+Ta) and Zr/Al_2O_3 vs. TiO_2/Al_2O_3 define volcanic-arc tectonic environment of their formation. MORB normalized trace elements patterns of the magmatic rocks (enrichment of large ion lithophile elements and low values for high field strength elements, the strong negative Ta and Nb anomaly indicate subduction related magma affinity. Chondrite-normalised REE patterns of the basaltic rocks confirm this conclusion. Nd and Sr whole rock isotope data reveal mantle-crustal signatures with ε-Nd (80 Ma) values generally between +2.1 and +3.0 and initial strontium ratios in the narrow range 0.704-0.705. Compared with the data for the Central Srednogorie the magmatic rocks from the Zidarovo ore field are less contaminated with crustal material. The same trend is observed comparing intrusive and volcanic varieties – the volcanic rocks reveal more primitive isotope composition, which is explained by the faster cooling of the volcanites, probably prior to contamination with crustal materials in middle/upper crustal chambers.

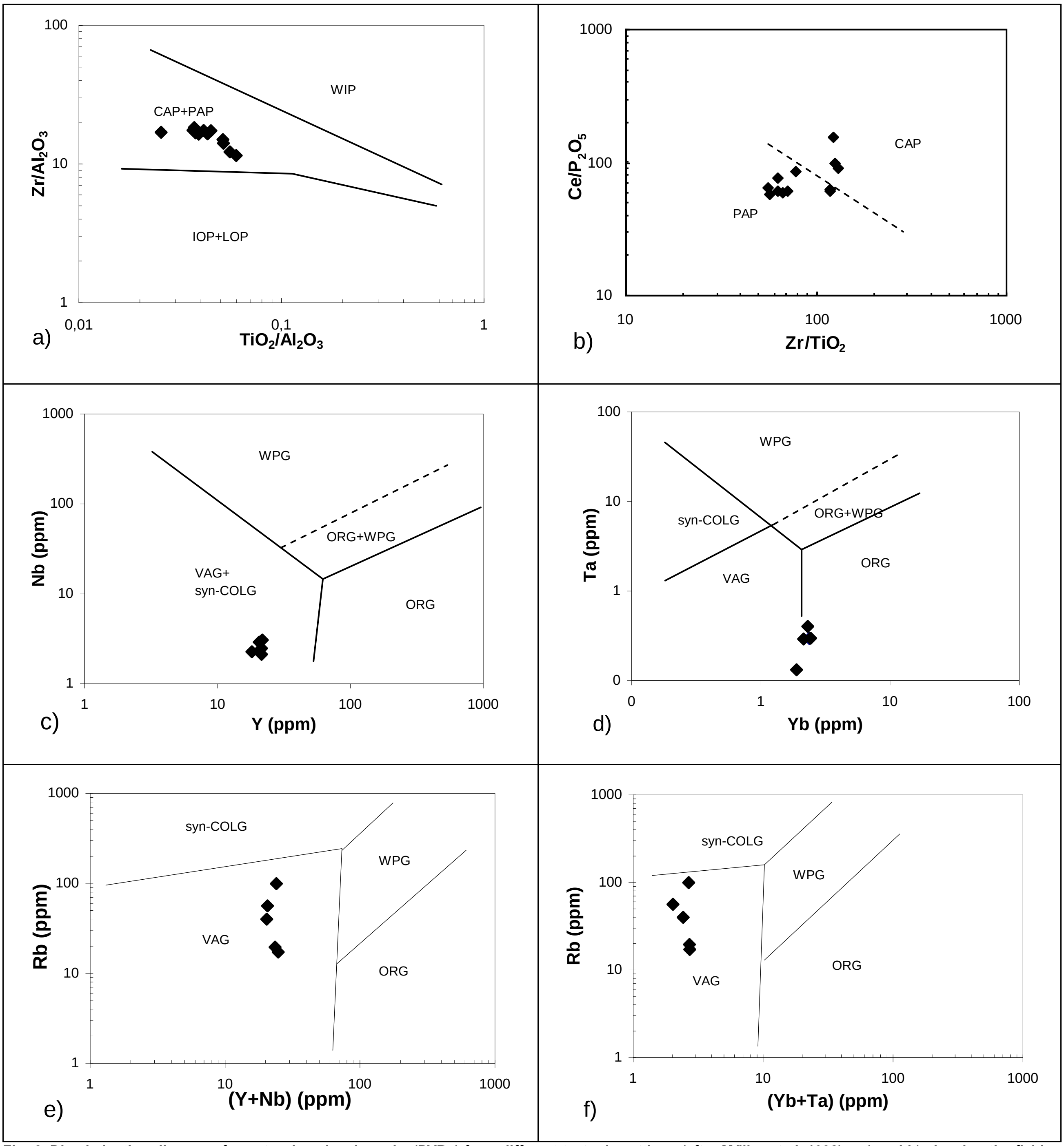

Fig. 6. Discrimination diagrams for potassic volcanic rocks (PVRs) from different tectonic settings (after Műller et al.,1992) – a) and b) showing the fields for within-plate PVRs (WIP), continental-arc (CAP) and continental/postcollisional PVRs (PAP), initial or late oceanic arc PVRs (IOP and LOP); Discrimination diagrams for granites (after Pearce et al., 1984) – c), d), e) and f), showing the fields of volcanic-arc granites (VAG), syn-collision granites (syn-COLG), within-plate granites (WPG) and ocean-ridge granites (ORG). a – Zr/Al_2O_3-TiO_2/Al_2O_3; b – Ce/P_2O_3-Zr/TiO_2; c – Nb-Y; d – Ta-Yb; e – Rb-(Y+Nb); f – Rb-(Yb+Ta)

Acknowledgements

This work was supported by the Swiss National Science Foundation through the SCOPES Joint Research Projects 7BUPJ62396, the Bulgarian National Science Foundation through the project NZ-1412 and by the SE Europe Geoscience Foundation. Present study is a contribution to the ABCD-GEODE (Geodynamics and Ore Deposits Evolution of the Alpine-Balkan-Carpathian-Dinaride Province) research program supported by the European Science Foundation.

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