COMPERATIVE PETROLOGY, GEOCHEMISTRY, Sr AND Nd ISOTOPE CHARACTERISTICS AND ORE GENETATION POTENTIAL OF THE LATE CRETACEOUS IGNEOUS ROCKS IN THE NORTHERN PART OF THE PANAGYURISHTE ORE REGION, SREDNOGORIE MAGMATIC ZONE

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ABSTRACT. The investigated Upper Cretaceous magmatic rocks are located in the region of the Chelopech Au-Cu epithermal deposit, the Zlatitsa pass and the Elatsite Cu-Au porphyry deposit. These igneous rocks are predominately of E-W orientation. They are mainly of andesitic, latitic and trachydacitic chemistry. They are of Turonian age according to radiogeochronological investigations of the dykes in the Elatzite deposit and the volcanics in the region of the Chelopech deposit. These magmatic rocks show similar petrographical, geochemical and isotopic composition and mineral chemistry. The main phenocrysts in the dykes and volcanics are represented by plagioclase and amphibole, rarely by quartz and biotite. The ground mass is microlitic. The trace element composition of these magmatic rocks is similar to those typical for the active continental margins (Andean type). Sr and Nd isotopic compositions suggest a mixed mantle and crustal source of the Turonian magma.

СРАВНИТЕЛНА ПЕТРОЛОГИЯ, ГЕОХИМИЯ, Sr И Nd ИЗОТОПНА ХАРАКТЕРИСТИКА И РУДОНОСНА ПЕРСПЕКТИВНОСТ НА КЪСНОКРЕДНИТЕ МАГМЕНИ СКАЛИ ОТ СЕВЕРНАТА ЧАСТ НА ПАНАГЮРСКИЯ РУДЕН РАЙОН, СРЕДНОГОРСКА МАГМЕНА ЗОНА

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РЕЗЮМЕ. Разглежданите горнокредни магмени скали се намират в района на находище Челопеч, Златишкия проход и находище Елаците. Тези скали са с пеобладаващо И-З разпространение. Те са с андезитов, латиов до трахидацитов състав. Възрастта им е вероятно туронска предвид датираните дайкови скали в района на нах. Елаците и Челопешките вулканити, които са в непосредствена близост и с подобен петрографски, геохимичен и изотопен състав както и минералин химизъм. Дайките са порфирни по плагиоклаз и амфибол, рядко кварц и биотит. Основната маса е микролитова. Микрохимичният състав на изследваните дайки е подобен на този, характерен за активните континентални окрайнини (Андийски тип). Съдържанието на Sr изотопи 0.7052 (коригирани за 90 млн. г.), а Nd изотопни отношения са 0.5124 (коригирани за 90 млн. г.).

Introduction

The aim of present investigation is to reconstruct the geological evolution of the Late Cretaceous Chelopech volcanic complex, dykes system eastern and north-eastern of it and the Elatzite subvolcanic bodies and porphyry dykes, and to identify the temporal relationships between its igneous products and other rock units and to outline the relation between magmatism and ore mineralization formation.

Geological setting and petrology of the investigated magmatic complexex

The products of the Late Cretaceous Chelopech volcanic complex, dykes system eastern and north-eastern of it and the Elatzite subvolcanic bodies and porphyry dykes are located in the Central Srednogorie volcanoplutonic area, which forms part of the Srednogorie tectonic zone (Dabovski et al., 2002; Fig. 1 and 2).

The basement of the igneous rocks consists of high-grade metamorphic rocks (two-mica migmatites with thin intercalations of amphibolites, amphibole-biotite and biotite gneisses), and low metamorphic phyllites and diabases of the Berkovitsa group (Early Paleozoic island-arc volcanic complex; Haydoutov, 2001). These units are in tectonic contact with each other, and to the North of Chelopech the phyllites of the Berkovitsa group are intruded by the Vejen pluton.

The Upper Cretaceous succession in the Northernmost part of the Panagyurishte ore region starts with conglomerates and coarsegrained sandstones intercalated with coal-bearing interbeds (coal-bearing formation; Moev, Antonov, 1978) covered by polymictic, argilleous and arkose sandstones to siltstones (sandstone formation). Collectively, these units have a thickness of less than 500 m. Pollen data suggests that both formations are Turonian (Stoykov, Pavlishina, 2003), where the age of 93.5 Ma was taken as transitional from the Cenomanian to the Turonian according to the Geological time scale. The sedimentary rocks are cut by volcanic bodies and overlain by sedimentary and volcanic rocks of the Chelopech Formation (Moev, Antonov, 1978). It comprises the products of the Chelopech volcanic complex, epiclastics, as well as the Vozdol sandstones. The latter ones are recently paleontologically dated as Turonian in age (Stoykov, Pavlishina, 2003). These formations have been eroded and transgressively covered by sedimentary rocks of the Upper Senonian Mirkovo Formation (reddish limestones and marls), which are in turn overlain by flysch of the Chugovo Formation (Campanian-Maastrichtian in age; Moev, Antonov, 1978).

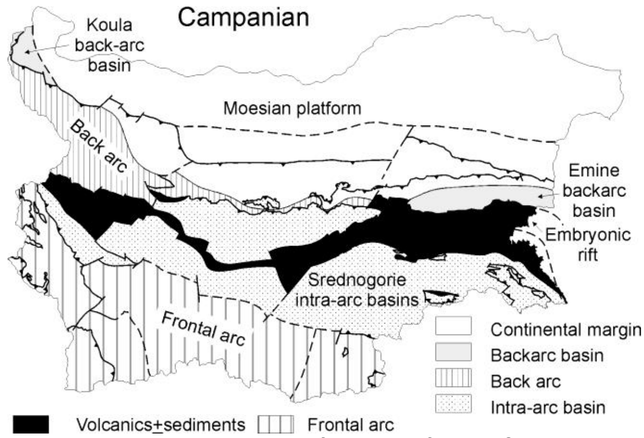

Fig. 1. Tectonic reconstruction of Bulgaria for the Campanian (Dabovski et al., 2002)

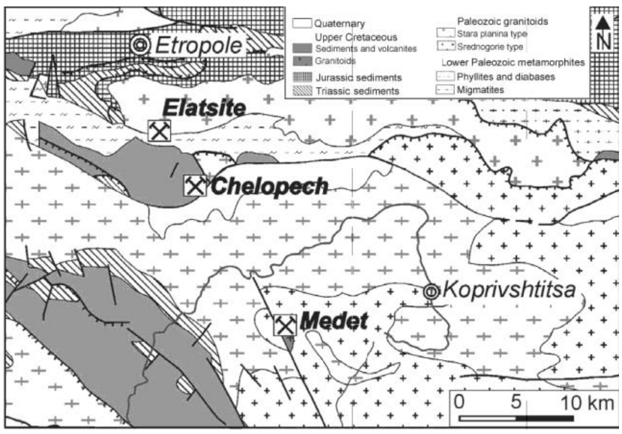

Fig. 2. Geological map of the Northern most part of the Panagyurishte region (modified by von Quadt et al., 2002, after Cheshitev et al., 1989)

Quartz-monzodiorite porphyries are presented in the region of the Elatsite deposit (Kamenov et al., 2003). They are the first and voluminous most important part of the ore-related group of dykes (included the sub-volcanic body called "the big dyke" in the deposit). Granodiorite porphyries (unit 2) and aplites (unit 3) completed the ore-related stage (von Quadt et al., 2002). The dykes from the post-ore mafic stage comprise unit 4 (microdiorite, micromonzodiorite, diorite porphyry and their quartz-bearing varieties) and unit 5 (quartz-diorite porphyry mainly). U-Pb geochronology revealed 92.3 Ma (unit 1), 91.84 (unit 2) and 91.42 Ma (unit 5, post-ore stage) (von Quadt et al., 2002). Ar-Ar age on biotite from the mine (Velichkova et al., 2001) of 90.5 Ma is interpreted as the closing time of the Rb-Sr and Ar-Ar isotope systems. A monzodiorite porphyry dyke yielded ages of 90.78 (amphibole) and 91.72 (biotite, Ar-Ar method; Handler et al., 2003). Rb-Sr isochron plot for the biotite and feldspar from rock unit 2 yields an age of 90.55 Ma (von Quadt et al., 2002).

Based on their structures, host rocks, crosscutting relationships and alterations Stoykov et al. (2002; 2003; 2004) divided the products of the Chelopech volcanic complex into 3 units: (I) dome-like volcanic bodies, (II) lava and agglomerate flows and (III) the Vozdol volcanic breccias and volcanites. The first unit is composed of dome-like volcanic bodies, which extruded through the unconsolidated Turonian sediments (the sandstone and coal-bearing formation) and through the metamorphic basement. The largest volcanic body (Murgana) is approximately 2x1 km in size. It shows higher stage of phenocryst crystallization than other units. Brecciated fragments of the dome-like volcanic bodies have been observed as xenoliths in the third unit of the Chelopech volcanic complex – the Vozdol volcanic breccia. The dome-like bodies mainly have an andesitic and trachydacitic composition. They are highly porphyritic (phenocrysts >40 vol.%). The phenocrysts consist of plagioclase, zoned amphibole, minor biotite, titanite and rare corroded quartz crystals, whereas the microlites consist of plagioclase and amphibole only. The accessory minerals are apatite, zircon, and Ti-magnetite. The second unit is represented by lava flows, which grade upwards into agglomerate flows (with fragments up to approximately 30 cm in size). Borehole data shows that the total thickness of these volcanic products is generally less than 1200 m, but exceeds more than 2000 m in the region of the Chelopech mine ("within their extrusive center"; Popov et al., 2002). The composition of the lava flows varies from latitic-trachydacitic to dacitic. Subsidiary andesites are also present. These volcanic rocks consist of the same phenocrysts, microlites and accessory minerals as the first unit, with the exception of the corroded quartz crystals. The lava flows contain fine-grained, fully crystallized enclaves of basaltic andesites to shoshonites. The enclaves consist of the same minerals as the main mass of unit 2 (plagioclase, amphibole and minor biotite), but comprise phenocrysts of different (more basic) chemistry (basaltic – andesite to andesite). A fine-grained quartz zone marks the margins of the enclaves. These features are typical for magma mingling and mixing processes and are mostly exhibited in the lava flows compared to the other volcanic units (Stoykov et al., 2002; Chambefort, Moritz, 2006).

Analytical techniques

Major and trace elements

Major and trace elements were analysed by X-ray fluorescence (XRF) at the University of Lausanne, Switzerland. Part was analyzed by ICP-atomic emission spectrometry in the University of Mining and Geology "St. Ivan Rilski". The rare earth elements (REE) were analysed by ICP-atomic emission spectrometry. The representative analyses of the compositional variation of the rock recovered from the studied volcanics are given in Tables 1 and 2. A petrological study has also been performed. Mineral analyses on samples of the different units were carried out at the University of Lausanne on a CAMEBAX SX-50 electron microprobe. Part 2 of these data are published in Stoykov et al. (2004; 2005).

Chemical composition of the igneous rocks

The magma of the Chelopech volcanic complex initially erupted more acid volcanic rocks. The earlier products (dome-like bodies and lava to agglomerate flows) contain 61-64 wt%

SiO_2 whereas the more basic Vozdol breccias and volcanites contained 55.5-58.0 wt% SiO_2.

Several dykes are exposed to the east- norteast compared to the Chelopech volcanic complex. They strike predominately in an east-west direction and intrude into the Pre-upper Cretaceous metamorphic basement. They do not show crosscutting relationship to the Chelopech volcanic complex.

The largest dyke is more than 7 km in length. These dykes have andesitic, latitic, dacitic and trachydacitic compositions. The phenocrysts consist of plagioclase, zoned amphibole, minor biotite, titanite and rare corroded quartz crystals, whereas the microlites consist of plagioclase and amphibole only.

Table 1
Major element composition of the representative samples (major components)

Oxides wt.%	Vozdol volcanics	Chelopech lava flows	Mafic inclusion	Dyke	Dyke
SiO_2	57.11	61.07	57.76	61.91	62.60
TiO_2	0.65	0.49	0.58	0.48	0.46
Al_2O_3	18.35	17.68	19.52	17.36	17.24
Fe_2O_3	7.03	4.56	6.8	4.45	4.24
MnO	0.12	0.13	0.08	0.14	0.15
MgO	1.75	1.49	1.14	1.26	1.22
CaO	4.87	4.9	3.14	3.09	4.35
Na_2O	4.19	4.21	6.01	3.78	4.07
K_2O	3.27	2.95	2.52	5.04	2.89
P_2O_5	0.26	0.22	0.25	0.22	0.19
LOI	1.55	1.54	1.94	2.21	2.82
Total	99.15	99.24	99.74	99.94	100.23

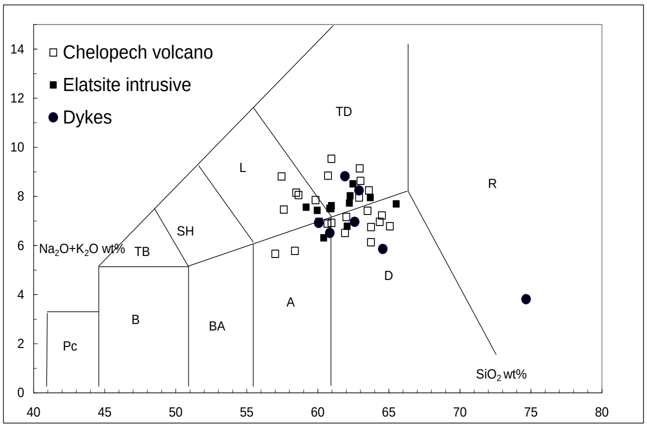

Fig. 3. TAS diagram after Le Maitre (1989) for representative samples from the studied region (B - basalt; BA - basaltic andesite; A - andesite; D - dacite; SH - shoshonite; L - latite; TD - trachydacite); data for the Chelopech volcanites are after Stoykov et al., 2004; for the dykes system eastern and north-eastern of the Chelopech volcano after Stoykov 2005; for the Elatzite after Stoykov & Popov (unpublished data)

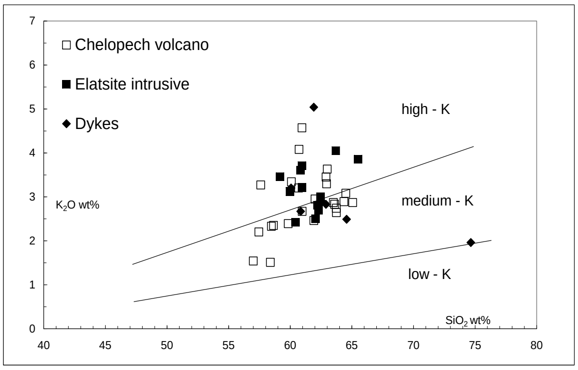

Fig. 4. SiO_2 vs. K_2O diagram after Le Maitre (1989) for representative samples. Data for the Chelopech volcanites are after Stoykov et al., 2004, for the dykes system eastern and north-eastern of the Chelopech volcano after Stoykov 2005, for the Elatzite after Stoykov & Popov (unpublished data)

Table 2
Major element composition of the representative samples (trace elements)

Elements (in ppm)	Dyke	Dyke	105GD	Chelopech lava flows	Vozdol volcanics
Nb	9	12	8	7	8
Zr	123	206	107	98	135
Y	22	27	22	20	22
Sr	641	794	1414	781	736
U	<2<	n.a.	<2<	n. a.	3
Rb	102	86	87	63	99
Th	4	n.a.	3	3	7
Pb	13	n.a.	15	16	21
Ga	19	18	18	19	18
Zn	49	81	45	72	52
Cu	7	n.a.	4	26	5
Ni	2	n.a.	2	2	3
Co	7	n.a.	39	10	25
Cr	13	n.a.	9	14	9
V	89	90	92	127	79
Ce	51	n. a.	41	49.3	43
Nd	25	n. a.	20	24	22
Ba	726	780	612	1441	771
S	11	n. a.	<3<	113	<3<
Hf	6	n. a.	7	6	6
Sc	10	n. a.	7	10	7
As	7	n. a.	7	6	4
La	25.20	n. a.	28.30	22.9	n.a.
Ce	53.30	n. a.	58.30	49.3	n.a.
Pr	6.40	n. a.	6.00	5.3	n.a.
Nd	24.80	n. a.	26.20	24	n.a.
Sm	4.90	n. a.	5.10	4.9	n.a.
Eu	1.23	n. a.	1.26	1.26	n.a.
Gd	3.60	n. a.	3.30	3.3	n.a.
Dy	3.20	n. a.	3.10	3.1	n.a.
Ho	0.67	n. a.	0.69	0.66	n.a.
Er	1.80	n. a.	1.80	1.8	n.a.
Tm	0.26	n. a.	0.28	0.26	n.a.
Yb	1.60	n. a.	1.70	1.5	n.a.
Lu	0.25	n. a.	0.25	0.22	n.a.

Mineral chemistry

The composition of plagioclase phenocrysts of the Murgana dome-like body of the Chelopech volcanic complex $An_{38.5-42.2}$ (core) to $An_{38.7-46.2}$ (rim); those of the lava flows varies from $An_{42.5-48.2}$ (core) to $An_{30.1-53.9}$ (rim); for the Vozdol volcanic rocks phenocrysts display range from center $An_{50.8}$ to $An_{36.2}$ in the periphery; and for dykes $An_{44.1-46.2}$ (core) to $An_{40.7-44.2}$ (rim). The rims are variable in composition and substantially overlap the field of the phenocryst cores the compositions of plagioclase microlites vary from An_{31} to An_{48}. K-feldspar microlites (Or_{86-93}) where only analyzed in the Vozdol volcanic rocks.

Table 3
The plagioclase composition of the representative samples

Sample	Pl/p.4l	Pl/p.5	Pl/p.6	Pl/p.3i	Pl/p.4c
SiO_2	58.30	58.82	58.59	58.16	58.06
TiO_2	0.05	n. d.	n. d.	n. d.	0.21
Al_2O_3	25.78	25.77	26.00	26.53	27.17
FeOtot	0.27	n. d.	0.07	0.11	0.24
MnO	n. d.	n. d.	n. d.	0.21	0.12
CaO	8.49	8.54	8.65	8.08	8.86
Na_2O	6.48	6.90	6.35	6.04	5.06
K_20	0.56	0.60	0.56	0.91	0.60
BaO	n. d.	n. d.	n. d.	n. d.	n. d.
Summe	99.93	100.63	100.22	100.04	100.32
Or	3.2	3.3	3.2	5.4	3.8
Ab	56.1	57.4	55.2	54.4	48.9
An	40.7	39.3	41.6	40.2	47.3

The composition of plagioclase phenocrysts (Table 3) of the investigated dykes north-eastern from the Chelopech volcanic complex varies from $An_{39.2-47.7}$ (core) to $An_{37.7-41.1}$(rim). The rims are variable in composition and substantially overlap the field of the phenocryst cores. The amphiboles display Mg# between 0.48 and 0.57. The contents of Si p.f.u. range between 6.40 and 6.48 and they plot on the limit of the magnesiohastingsite, and hastingsite field of Leake et al. (1997).

The amphiboles for all volcanic rocks display $Mg^{\#}$ between 0.48 and 0.67. The contents of Si p.f.u. range between 6.40 and 6.55 and they plot on the limit of the magnesiohastingsite, pargasite, ferropargasite, hastingsite and Fe-edenite field of Leake et al. (1997). The composition of the amphibole crystals of the inclusions is different to the one of the volcanic rocks. It displays higher values of Mg# between 0.70 and 0.83 and is classified as magnesiohastingsite. The contents of Si p. f. u. of the amphiboles from the inclusions range between 5.90 and 6.10.

Bulk rock trace elements composition

The MORB normalized patterns for the investigated magmatic rokcs (Table 1 and 2; Fig. 5) indicate enrichment of LILE and in lesser degree of some HFSE (Ce, Zr, P and Hf) with a strong negative Nb anomaly and a depletion of the Fe-Mg elements. All these features are typical for subduction-related magmatic sequences due to the melting of sedimentary material of the subducted slab. In comparison to the volcanic rocks of an Andean-type active continental margin, the investigated magmatic rocks show small K_2O, Ba and Hf enrichments and depletions of Nb, TiO_2, Zr and P_2O_5. The Elatzite igneous rocks show relatively increased Rb, Th and Cr ratios and decreased P, Zr, Hf, Ti, Y and Sc ones.

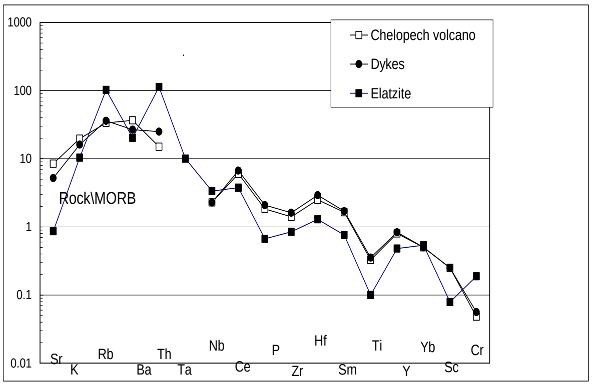

Fig. 5. Spider discrimination plot for the investigated dykes. Data for the Chelopech volcanites are after Stoykov et al., 2004; for the dykes system eastern and north-eastern of the Chelopech volcano after Stoykov 2005; for the Elatzite after Kamenov et al., 2003

These rocks have fractionated LREE and relatively flat HREE patterns (Table 2, Fig. 6), as typically found in subduction related volcanic rocks. The LREE enrichment ranges from 33 to 80 times chondritic, whereas La_n/Yb_n ratios vary from 10 to 13. Middle and heavy REE show relatively flat patterns, generally within 5-30 times that of chondritic ones. An Eu anomaly is not observed, which suggests that there was no plagioclase fractionation involved in genesis of the studied andesitic rocks. The Elatzite igneous rocks show relatively increased Gd, Dy, Ho, Eu, Tm, Yb and Lu ratoes.

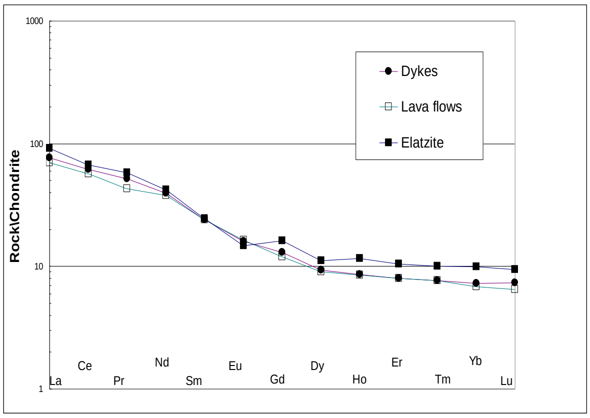

Fig. 6. Rock/chondrite diagram for the investigated magmatic rocks. Data for the Chelopech volcanites are after Stoykov et al., 2004, for the dykes system eastern and north-eastern of the Chelopech volcano after Stoykov 2005, for the Elatzite after Kamenov et al., 2003

Sr and Nd isotopes

Rb-Sr and Sm-Nd whole rock isotope analyses

The isotopic composition of Sr and Nd and the determination of Rb, Sr, Sm and Nd contents were performed at the University of Geneva. The initial Sr ratios for the investigates dyke complex range between 0.7055 and 0.7060 (after 90 Ma correction). The Sr isotope ratios of the magmatic rocks from the Chelopech volcano display a small range between 0.7049 and 0.7054 after a 90 Ma correction (Stoykov et at., 2002). Generally $^{87}Sr/^{86}Sr$ ratios fall within the field previously defined by Kouzmanov et al. (2001) values from 0.7046 to 0.7061 (after 80 Ma correction) for the volcanic (andesites and dacites) and plutonic (granodiorites and granites) rocks from the southern part of the Central Srednogorie volcano-intrusive area. The Nd isotope ratio for the investigated dykes from the Zlatiza pass varies from 0.512449 to 0.512450 (after 90 Ma correction). The calculated ε90(Nd) values are between -2.27 and -3.55. These data are similar to Sr and Nd isotope composition of the Chelopech volcanites (Stoykov et al., 2004) and the Elatsite subvolcanic rocks. They suggest a mixed mantle-crust source of the Turonian magmatism in the Chelopech region. However using the variations of the initial Sr and Nd ratios vs. SiO_2 the evolution of the magma may be largely due to mingling/mixing processes, without isotope homogenisation in the whole volume of the magma chamber, and not to a simple differentiation of one parental magma, combined or not with assimilation of upper crustal rocks (Stoykov et al., 2004).

Conclusions

The investigated Chelopech volcanites and dykes from the Zlatiza pass are of andesitic, latitic to dacitic and trachydacitic chemistry. Their phenocrysts (>40 vol.%) consist of plagioclase, zoned amphibole, minor biotite, and titanite; whereas the microlites consist of plagioclase and amphibole only.

The MORB normalized patterns for the mamatites form the Northern part of the Panagyurishte ore region indicate enrichment of LILE and in lesser degree of some HFSE (Ce, Zr, P and Hf) with a strong negative Nb anomaly and a depletion of the Fe-Mg elements. All these features are typical for active continental margin. These rocks have fractionated LREE and relatively flat HREE patterns, as typically found in subduction related volcanic rocks. The LREE enrichment ranges from 33 to 80 times chondritic, whereas La_n/Yb_n ratios vary from 10 to 13. Middle and heavy REE show relatively flat patterns, generally within 5-30 times that of chondritic ones. An Eu anomaly is not observed, which suggests that there was no plagioclase fractionation involved in genesis of the studied andesitic rocks.

The initial Sr ratios for the Chelopech volcanites and dykes from the Zlatiza pass complex range between 0.70550 and 0.70601 (after 90 Ma correction) and the Nd isotope ratio varies from 0.512449 to 0.512450.

The combined petrologic, isotope-geochemical and geochronological investigations of the dyke complex in the Zlatitsa pass suggest similar composition compared to the Chelopech volcanites and the Elatsite subvolcanic rocks.

The petrological and geochemical features give additional evidence for a possible uniform magma chamber of the volcanic rocks in the Chelopech and Elatsite deposits (Stoykov et al., 2004) and the investigated dyke complex in the Zlatitsa pass with a complex evolution in Turonian times, when a combination of processes of magmatic differentiation, assimilation, mingling and mixing took place. These magmatic products reveal similar Sr and Nd characteristics (Stoykov et al., 2002; von Quadt et al., 2002), where the tendency of an increase of initial Sr and Nd isotope ratios related to minor assimilation of host rocks within parts of the magmatic chamber. The amphibole chemistry of the magmatic units of both deposits and the investigated dyke complex shows some similar characteristics – Mg# between 0.48 and 0.67 and Si per formula unit content between 6.40 and 6.55, but mark differences comparing to the other deposits of the Panagyurishte ore region (Stoykov et al., 2002; Kamenov et al., 2003).

The porphiry-copper Elatzite and the copper-gold epithermal Chelopech deposits are related to described Late Cretaceous subvolcanic and volcanic magmatic products. Quartz-monzodiorite porphyries (unit 1) are the first and voluminously most important part of the early ore-related group of dykes (includes the subvolcanic body called "the big dyke" in the Elatzite deposit). Granodiorite porphyries (unit 2) and aplites (unit 3) completed the ore-related stage (von Quadt et al., 2002). The ore mineralization in the Chelopech deposit is hosted in the second stage of the volcano (lava flows, Stoykov et al., 2004).

Ore mineralizsations related to the dyke complex in the Zlatitsa pass are not described.

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