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STEFAN BOYADJIEV*

GEOCHEMISTRY OF RARE-EARTH ELEMENTS IN MIDDLE ALPINE PLUTONS FROM THE CENTRAL SREDNOGORIE, BULGARIA

(Tabs. 1, Figs. 2)

Abstract: The complex plutons studied show a constant deficiency in $\sum$TR — for intermediate and acid rock varieties, and resemblence to tholeiites — for gabbros. All rocks exibit Ce character of the lanthanoids and negative Eu anomaly. In the process of fractionation the potentially metal-bearing plutons reveal a tendency to relative concentration of intermediate and heavy lanthanoids. $\sum$TR in the plutons of the two petrochemical trends mark a single genetic code which defines their comagmatic links. The primary, Ca-alkaline magma type was probably formed at a crustal-mantle level.

Резюме: Рассматриваемые комплексные плутоны характеризуются постоянным дефицитом $\sum$TR для среднеосновных и кислых разновидностей пород, и сходством с толеитами для габбро. Все породы отличаются цериевым характером лантаноидов и отрицательной европиевой аномалией. В процессе фракционирования, потенциально металоносные плутоны показывают тендецию к относительному обогащению средными и тяжелыми лантаноидами. $\sum$TR для плутонов двух петрохимических рядов (трендов) намечает единный генетический код, что определяет их комагматичность. Первичная магма кальциево-щелочного типа вероятно формировалась на корово-мантийном уровне.

Introduction

The studies of rare-earth elements (REE) in the magmatic formations of Bulgaria, particularly in the Middle Alpine plutons, date back to about 15 years ago. As a result some more important features in their behaviour during the magmatic process have been revealed. A relative deficiency of ΣTR and dominant cerium character of the lanthanoids has been traced. Certain correlation link between ΣTR and the total alkalinity of the subalkaline plutons (Vitosha, Rosen) has been established. The main form of occurrence of REE is isomorphic dispersion in the main rock-forming minerals and subordinate amounts in own accessory minerals (Aleksiev et al., 1967, 1969; Aleksiev, 1969, 1974). On the background of a negative anomaly the values of Eu decrease parallel to the general acidification of the magmatic derivatives (Daieva, 1980).

The purpose of the present paper is, on the basis of a considerably larger number of samples analysed by means of modern methods and precise equipment, to study in more detail the geochemistry of the rock varieties in the investigated plutons, to determine the path of magmatic differentiation and the related lanthanoid fractionation and eventually to evaluate the depth of generation of the respective magmas.

*Prof. St. Boyadjiev, Geological Institute, Bulgarian Academy of Sciences, Acad. G. Bončev Str., block 24, 1113 Sofia.

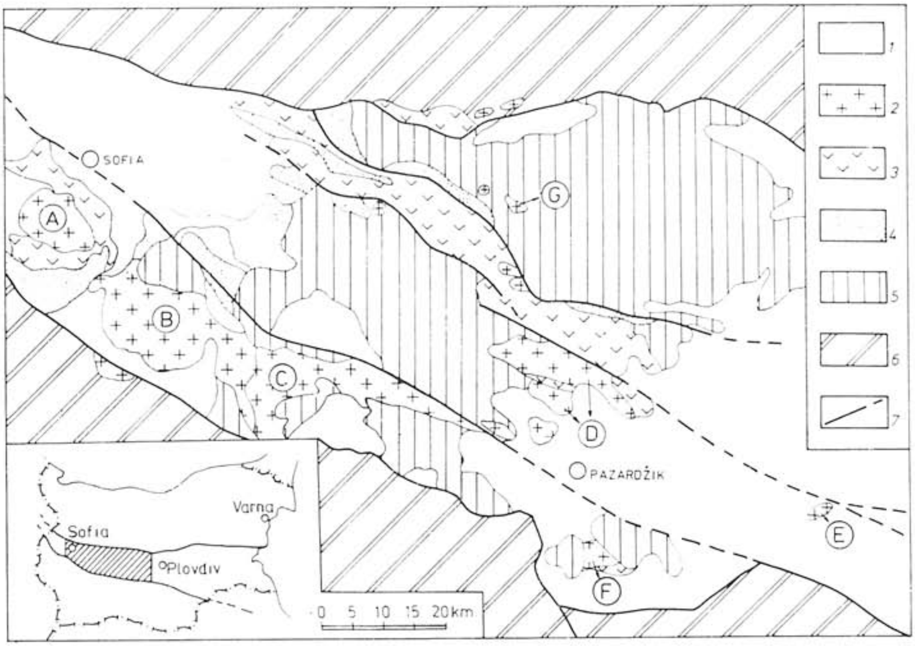

Fig. 1. Geological map of a part of the Central Srednogorie.
Explanations: 1 — Neozoic sedimentary complexes; 2 — Middle Alpine plutons: Vitosha (A), Plana (B), Gutsal (C), Elshitsa-Boshulja (D), Plovdiv (E), Kapitandimitrievo (F), Medet (G), framework of the plutons; 3 — Upper Cretaceous volcanogenic-sedimentary complex; 4 — Paleozoic-Mesozoic rock complexes; 5 — granitoid-consolidated Precambrian crystalline basement; 6 — structural zones bounding the Central Srednogorie; 7 — faults.

The studies cover representatives of the two petrochemical (Ca-alkaline and K-subalkaline) trends. Seven intrusives from the Central Srednogorie and individual representatives of the accompanying dike formation were studied (Fig. 1). A total of 71 samples was analysed by means of neutron-activation analysis (NAA). The analysis was carried out by Cubriev and Matanov to whom I express my deep gratitude.

All samples were analysed for La, Ce, Nd, Sm, Eu, Tb, Yb, Lu, the maximum error being below 10 %.

The results from single samples were grouped according to facies and recalculated into "average" values which eliminate eventual extreme values and yield most accurate data for individual elements in the rock type as a whole (Table 1). In the computation of the average data results from former publications are included (Aleksiev et al., 1964, 1969; Aleksiev, 1974; Daieva, 1980).

In order to elucidate the degree of fractionation of REE the obtained average data are chondrite normalised (according to contents given by Massuda et al., 1973). The graphs of REE distribution (Fig. 2) are constructed on this basis.

Brief petrological description of the plutons

The Middle Alpine (Neointrusive, Late Hercynian, Laramide) intrusive magmatism in the Central Srednogorie is represented by Vitosha (Dimitrov, 1942), Plana, Gutsal, Plovdiv (Boyadjiev, 1962, 1971, 1973), Elshitsa-Boshulja (Boyadjiev — Cipčakova, 1964), Kapitandimitrievo (Boyadjiev — Panayotova, 1982) and Medet (Ušev et al., 1962) plutons (Fig. 1). Except for Gutsal and Plovdiv pluton the other bodies are of composite, polyfacies structure. Their facies development occurred in three main stages.

According to the results of the former studies the basic, mainly gabbro intrusion, was formed during the first stage. As a result dominantly of gravity differentiation anorthosites and pyroxenites are locally found in certain cases. In the form of relatively small, chain-like bodies the gabbro was intruded along a petro-lineament and first marks the Middle Alpine intrusive activity in this region. Commonly the gabbro bodies are disrupted and form xenoliths in the rock varieties of the next magmatic impulses. This leads to formation of hybrid facies of transitional character: monzogabbro in Vitosha, gabbrodiorites, granogabbro, etc. in Plana pluton. These rock varieties show higher content of K_2O.

During the second, main intrusive stage, as a result of homodromous development, the next, intermediate to acid rock varieties were formed. Two models of polyfacies plutons may be distinguished: differentiation and crystallization "in situ" at hypabyssal levels (Plana, Kapitandimitrievo, etc.), and abyssal differentiation with intrusion and crystallization of the magmatic portions at higher levels (Vitosha).

Most intrusive bodies are cut by vein rocks formed during the third, relatively independent stage. Usually, their facies and mineral composition is analogous to that of the representatives of the second magmatic stage. A poor aplite-pegmatite phase is related to the plutonic bodies. It contains a rich mineral association including minerals, concentrators of REE — orthite, monazite, uranothorite (Boyadjiev — Ivanov, 1975, 1982).

The Middle Alpine intrusive magmatism in the Central Srednogorie, and in the Srednogorie zone in general, terminates with the so-called "Srednogorie dike formation" (Dimitrov, 1959). It is represented by two series which are relatively independed in temporal and petrochemical respect intermediate to acid derivatives of the Ca- and K-alkaline petrochemical series and a relatively later lamprophyric association (Boyadjiev, 1979).

In petrochemical respect the Vitosha pluton is a representative of the Mediterranean tendency (Dimitrov, 1942) while the other intrusives bear the features of the Ca-alkaline series; in them the alkalization overtakes the acidification.

Distribution of rare-earth elements in the rock varieties of the plutonic bodies

In accord with the large petrographic variations, ΣTR in the plutons under consideration ranges from 46 to 164 ppm and only in Plovdiv pluton reaches 235 ppm (Table 1). The following main features are typical of individual plutons and their rock varieties.

Table 1

The rare-earth elements content in the rock types of Middle — Alpine intrusions from Central Srednogorie, Bulgaria (in ppm)

Pluton	Rock (from ... nuber specimens)	La	Ce	Nd	Sm	Eu	Tb	Yb	Lu	$\sum TR$***	La/Yb	Ce/Yb	$\sum Ce$***/$\sum Y$	Eu
Vitosha	Gabbro (4)	23.6	51.0	13	4.7	0.92	1.20	1.9	0.37	117.5	11.7	26.8	6.5	0.6
	Monzonites (7)	18.0	33.1	25	4.4	0.95	1.03	1.8	0.17	104.5	9.8	18.7	6 2	0.7
	Sienites (4)	25.4	49.2	29	4.5	0.82	0.85	2.2	0.42	132.0	12.2	23.5	8.1	0.7
	Vein granosie-nites (2)	26.5	50.0	30	4.9	0.38	0.97	2.4	0.34	135.5	11.2	21.1	7.7	0.3
Plana	Gabbro (3)	14.8	31.7	29	4.0	0.90	1.00	1.7	—	102.7	8.7	18.6	5.8	0.7
	Diorites (6)	29.7	61.1	16	5.8	**0.60**	1.15	2.4	0.15	160.7	13.3	27.0	6.3	0.6
	Quartz-monzo-diorites (4)	22.5	47.4	25	5.4	0.55	1.05	2.4	0.30	124.7	11.5	22.8	5.6	0.7
	Granodiorites (4)	32.7	66.7	20	4.9	0.45	0.52	1.2	0.00	164.1	26.3	52.7	13.4	0.7
	Adamelites (2)	33.9	59.5	30	6.4	1.00	0.95	1.7	0.35	142.9	11.0	19.6	8.3	0.5
	Vein granites (2)	26.5	47.0	29	7.4	1.00	1.20	2.4	0.20	136.9	19.4	19.1	6.6	0.6
Gutsal	Granodiorites (5)	36.4	77.0	9	6.0	0.90	0.94	2.1	0.18	158.5	19.2	40.3	8.6	0.6
Elshitsa-Boshulya	Gabbro (4)	10.9	15.2	2	3.5	0.67	0.27	1.2	0.17	45.8	8.8	12.4	3.6	0.7
	Granites (3)	33.2	32.3	18*	3.9	0.53	0.43	1.9	0.10	97.7	17.0	16.6	16.9	0.6
	Vein granites (2)	33.2	48.5	15*	2.6	0.40	0.35	0.6	0.20	112.0	51.1	74 7	15.5	0.7
Kapitan-dimitrievo	Gabbro (3)	11.7	24.3	—	6.7	0.97	1.17	1.6	0.31	64.80	7.3	15.2	2.0	0.5
	-Diorites (3)	18.8	29.3	—	6.3	0.90	1.10	1.5	0.17	75.10	19.6	19.6	2.8	0.5
	Quartz-monzo-diorites (4)	34.3	53.2	3.5	6.1	1.12	1.05	1.9	0.26	117.80	27.7	27.7	3 9	0.6
	Granodiorites (8)	21.6	38.1	20.0	5.0	0.74	0.80	1.5	0.24	102.90	24.6	24.6	5.2	0.5

Medet	Quartz-monzo-diorites (2)	40.2	75.5	13	6.3	0.85	1.05	1.9	0.25	159.0	21.2	39.7	9.6	0.5
	Granodiorites (3)	37.2	50.3	44	6.9	0.73	0.83	1.5	0.17	161.1	25.3	34.2	12.2	0.5
	Vein granitoides (2)	9.0	17.5	15	2.3	0.65	0.35	0.8	0.15	54.7	10.6	20.6	8.1	0.9
Plovdiv	Granodiorites (2)	39.1	78.0	82	7.2	1.40	0.80	1.6	0.10	234.9	24.4	48.7	18.9	0.7
	Vein granodiorites (3)	25.2	42.7	19	3.8	1.00	0.63	1.3	0.18	107.5	19.8	33.6	10.4	0.9
Dike formation	Diorite-porphyrites (1)	17.8	53.0	19*	3.3	1.20	0.60	1.6	0.30	112.1	11.1	33.1	9.6	1.0
	Granodiorite porphyrites (1)	23.5	54.0	24*	4.5	0.90	0.90	2.2	0.10	129.4	10.7	24.5	7.7	0.7
	Granite-porphyrites (1)	17.3	34.0	15*	2.8	0.30	0.50	1.7	0.20	83.3	10.2	20.0	7.4	0.4
	Lamprophyres (2)	13.0	30.0	17*	4.1	0.95	1.20	1.6	0.15	87.6	7.9	18.5	4.6	0.7

* The data for Nd are derived from the chondrite curve.

** $\sum TR$ is computed on the basis of data for the indicated elements including the content of the other lanthanoids which are derived from the chondrite curve.

*** $\sum Y$ is without itrium content which is not analyzed for methodical reasons.

1. Different ΣTR in rock varieties of one and the same type from different plutons is established. Evidently, this is a result of the relatively independent development of each intrusive irrespective of their common comagmatic and formational links. The ruptural predestination of the magmatic chambers, the formation of individual partial magmas as well as the particular conditions of their differentiation and crystallization govern the specific conditions of the rare-earth fractionation.

2. According to ΣTR content the gabbro impulses, with certain quantitative variations, show similarity to island-arc (57 ppm) and continental (91 ppm) tholeiites (after Balašov, 1976). They are also relatively rich in light lanthanoids. ΣTR is relatively higher in the gabbro from Vitosha and Plana. In this case, however, there is a remarkable correlation with the higher K_2O content (1.01 and 1.38 respectively) which was evidently supplied by the later magmatic portions.

In the diagrams showing the lanthanoid fractionation (Fig. 2) and particularly in those which trace the internal distribution of REE in the process of magmatic differentiation the gabbro has a specific position with respect to the other rock varieties from Elshitsa-Boshulja pluton (Fig. 2 — B); in Vitosha and Plana intrusives this feature is masked by the above mentioned assimilation and hybridization processes. The lamprophyres of the dike formation show analogous behaviour (Fig. 2 — C). This suggests that the magmas which formed the basic rock varieties of the composite plutons and the lamprophyres of the dike formation were relatively independent both in temporal and petrochemical respect and maybe also in genetic respect.

3. All other rock varieties of the second and third magmatic stage of the two petrochemical series (except for Plovdiv pluton) show a subclark content of ΣTR and distinct deficiency in respect of the equivalent average rock types (after Balašov). At the same time the ΣTR content in the subalkaline varieties of Vitosha pluton and the intermediate to acid derivatives of the intrusives of the Ca-alkaline series are relatively equal. This suggests that the magmas of the K-subalkaline plutons in the Srednogorie zone were probably formed as a result of abyssal differentiation of magma of primary Ca-alkaline composition.

On the basis of the relatively higher ΣTR in Medet pluton Aleksiev (1969) assumed certain relations to its potential metal content. In this respect there are certain analogies with Plana, Gucal and Plovdiv pluton (Table 1) where some hydrothermal mineralizations are also found.

4. In the rock varieties of the second magmatic stage in Vitosha pluton the normal order of lanthanoid fractionation established by Aleksiev et al. (1964) and Daieva (1980) is confirmed: the ΣTR content gradually increases towards the later and relatively more acid varieties. There is also a distinct relation between the degree of REE fractionation and the development of magmatic differentiation in view of the gradual increase of the total and mainly of the K-alkalinity; at the same time the relation between CaO and fm is reverse and is due to the considerable decrease of coloured minerals.

However, this tendency is unstable for the rock varieties of the second main magmatic stage of the Ca-alkaline series. In Medet pluton it is not well expressed, in Plana shows intrafacial variations and in Kapitandimitrievo intrusive it is not expressed at all. On the background of a general tendency for a slight but gradual increase of the total alkalinity a partial variation of the behaviour of K_2O in the differentiation process is commonly observed. There is also certain relation to the content of hornblende and in parts of biotite.

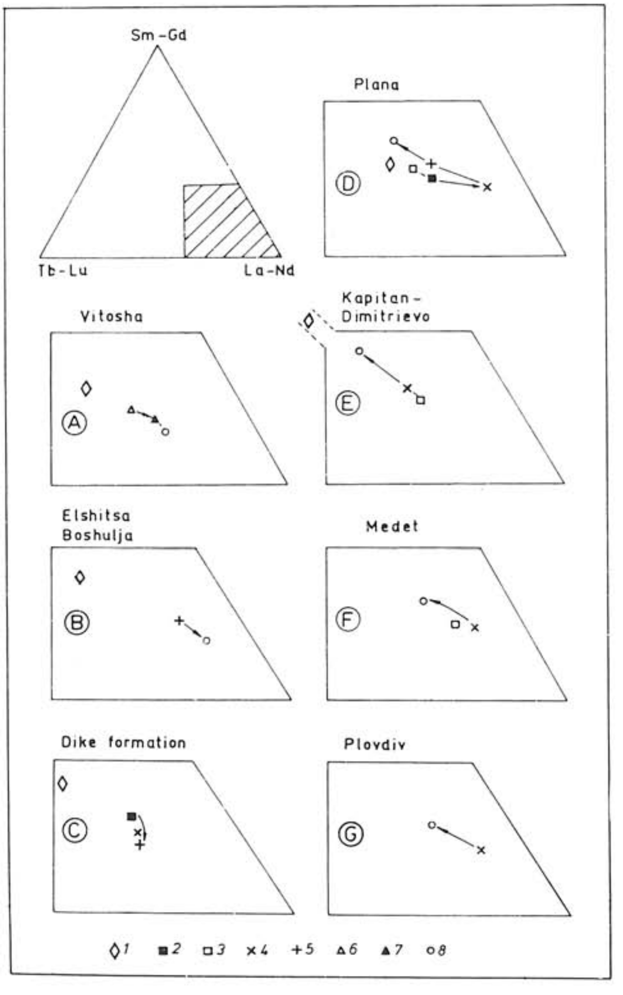

Fig. 2. Diagrams showing the fractionation models of light (La—Nd), intermediate (Sm—Gd) and heavy (Tb—Lu) lanthanoids in some of the Middle Alpine plutons from Central Srednogorie.

Explanations: A — Vitosha pluton, B — Elshitsa-Boshulja pluton, C — dike formation, D — Plana pluton, E — Kapitandimitrievo pluton, F — Medet pluton, G — Plovdiv pluton, rock varieties: 1 — gabbro; 2 — diorite; 3 — monzodiorite; 4 — granodiorite; 5 — adamelite, granite; 6 — monzonite; 7 — syenite; 8 — vein granitoids.

In the group of vein granitoids of the third magmatic stage two tendencies in the behaviour of ΣTR are recorded. In Vitosha, Plana and Elshitsa-Boshulja pluton, the general regularity of the second stage is still operating — gradual increase towards the end facial members. This fact indicates that they are formed as a result of a single differentiation series irrespective of their relative temporal and lateral independence. In Medet and Plovdiv pluton, however, the vein granitoids show significantly lower ΣTR content which suggests that they are better individualized in temporal and possibly in genetic respect. This is confirmed by the fact that the vein rocks which are implaced in the Medet pluton (sulphide mineralization of areal type) are "barren". This suggest a relatively independent development of the vein rocks.

On the basis of isolated data (Table 1) certain instability of ΣTR is traced also in the dike formation. The behaviour of ΣTR here may be explained by the relatively independent development of the individual magmatic portions which were differentiated at abyssal level and were temporally and laterally individualized.

5. The variations in the degree of REE fractionation in some of the plutonic representatives is of definite interest. In the subalkaline Vitosha pluton and in the oversaturated in SiO_2 Elshitsa-Boshulja pluton as well as in the dike formation (of the Ca-alkaline series) the fractionation is of "one-way" type, showing a tendency to an uninterrupted increase of light lanthanoids (Fig. 2 — A, B, C). This co-variant model is assumed as a criterion for the development of fractional differentiation in the comagmatic series (Aleksiev, 1969; Balašov, 1976; Daieva, 1980). However, in part of the plutons under consideration and particularly in the largest and polyphase Plana pluton (Fig. 2 — D), there is a distinct inversion in the fractionation during the formation of the transitional granodiorite facies. In this intermediate link of the general facial series the content of ΣTR reaches a maximum and after that it decreases. The part of the intermediate and heavy lanthanoids increases correspondingly, mainly at the expense of the considerably decreased content of Ce (Table 1). This irregular distribution of REE in Plana pluton is explained by Aleksiev et al. (1969) with influence of contamination processes. However, as far as such processes have not been proved (Boyadjiev, 1971) this inversion most probably may be related to the following petrochemical-mineralogical features: the higher ΣTR content in the diorite depends evidently on the relatively higher amount of hornblende (as an active concentrator of REE) and in the granodiorite — on the high K_2O content; in the quartzmonzodiorite where ΣTR is relatively lowest both hornblende and K_2O show a relatively lower content.

Consequently, it may be assumed that ΣTR in the plutons discussed is controlled by a complex of factors: the general petrochemical character of the initial — producing magma, the conditions of its deep fractionation and respectively the specific features in the formation of the partial magmas, the conditions of crystallization differentiation in the respective magma chambers, an important factor is also the quantitative and qualitative composition of the rock-forming components — as concentrators of REE in the particular rock varieties.

6. All rock varieties of the two petrochemical trends are characterized by a distinctly expressed cerium character of ΣTR which is well demonstrated

by the ratio $\Sigma Ce/\Sigma Y$. A particular relation to the facies conditions, however, is not established. The variation of ΣTR in the rock varieties of the individual plutons depends on the content of Ce and in Medet and Plovdiv plutons — also on the content of Nd (Table 1).

On the background of the distinct Ce character of ΣTR in Medet, and Plovdiv pluton the processes of REE fractionation in the development of magmatic differentiation and crystallization show a tendency towards a gradual increase of intermediate and heavy lanthanoids. Similar processes are traced in the later stages also for Plana pluton (Fig. 2 — D, F, G). All these plutons show a higher metallogenic potential.

The ratio $\Sigma Ce/\Sigma Y$ confirms the opinion of Aleksiev (1969) on the relatively higher mobility of the cerium group and on the development of two relatively independent tendencies in REE fractionation. Higher itrium content is recorded in the K-subalkaline Vitosha pluton and in parts in the intrusives of the Ca-alkaline group (Plana, Kapitandimitrievo) which show a relatively higher potash tendency. The predominant part of the plutons with distinct pacific tendency is characterized by higher cerium content.

The fact that the dike formation falls within the group of higher itrium content suggests that, irrespective of the noticable links with some plutonic bodies and their probable comagmatic origin, it developed geochemically as independent unit. This confirms its relative temporal and lateral individuality.

7. The ratio La/Yb likewise indicates the predominance of light lanthanoids in the process of fractionation. Again, two relatively independent groups of plutons may be distinguished. The first group includes the subalkaline intrusives as well as representatives of the dike formation. Here, the above ratio varies between 8 and 12. This analogous behaviour probably depends on the higher alkalinity of the aschistic group of the Ca-alkaline series. The second group, where the ratio La/Yb is 11 to 51, covers all rock varieties of the second and third magmatic stages of the Ca-alkaline series (Table 1). In accord with the concept of Lutz (1974) on the limited variations and the average value of La/Yb it may be assumed that the initial magmas of the two petrochemical groups were generated at a level of 50—100 km and more. The deep, crustal-mantle genesis of the magma is witnessed also by its petrochemical and metallogenic features: relatively higher content of MgO, Fe_2O_3, CaO and above clark backround of Cu, Mo, in parts Pb and Zn (Boyadjiev, 1979).

8. In most rock varieties of the studied plutons the content of Eu is in the limits of island-arc tholeiites (Balašov, 1976). This demonstrative fact likewise supports the idea on the deep, mantle genesis of the primary magmas (Boyadjiev, 1979).

The tendency towards a gradual decrease of the relative concentration of Eu in the end facies members in the subalkaline Vitosha and Rosen plutons (Daieva, 1980) may be traced also in the rock varieties of the second magmatic stage for the plutons of the Ca-alkaline series. This trend is very distinct in the intermediate and acid derivatives of the dike formation (Table 1). Thus, there is a reverse correlative link between the gradual acidification and the nominal quantity of Eu. The increasing deficiency of Eu in the end members of the magmatic series is a characteristic criterion for the development of fractional differentiation in comagmatic series as pointed out by Balašov (1976) and Daieva (1980).

The lamprophyres, as end members of the dike formation, show higher content of Eu as compared to the preceeding granite porphyries. This indicates that, irrespective of the assumed comagmatic origin, the basic partial magmas have a relative independent geochemical development during their abyssal evolution. The reasons for the considerable quantitative differences in the ΣTR content of gabbroids and the following facies varieties are probably very similar. In this light the idea of Dimitrova et al. (1983) that the rock varieties of the first (gabbroic) and the next magmatic stages in the polyfacies plutons belong to one and the same formation is evidently not well grounded.

All rock varieties of the intrusive bodies under consideration, as well as those of the dike formation, show a distinct negative Eu anomaly which may be assumed as a criterion for their primary magmatic genesis.

The parallel decrease of the Eu values towards the end and more acid members of the K-subalkaline magmas (Daieva, 1980) has not been observed in the Ca-alkaline series. At the same time there are certain differences in the Eu values between the end members of the second magmatic stage and the vein granitoids (Table 1). Constant differences are observed also in the Eu values of gabbros and the rock varieties of the second magmatic stage for the two petrochemical series which usually show identical values. Distinct differences are evident also for different varieties of the dike formation. In all three cases, however, a co-variant petrochemical relation is not established. This internal facies relation in lanthanoid fractionation probably reflects their relative petrochemical individuality which is indirectly expressed in their temporal and spatial independence.

Conclusions

The distribution of REE in the rock varieties of the Middle Alpine plutons leads to the following more important conclusions:

1. A relative deficiency of ΣTR in respect of the average rock types has been established. The ΣTR content in the basic, gabbroid impulse as well as the general subclark content in all rock varieties of the second and third magmatic stage indicate a tholeiitic character of the generating magmas.

The considerable deficiency of ΣTR in the subalkaline Vitosha pluton as well as its close content in the rock varieties of the Ca-alkaline series suggests that they were formed in the process of deep differentiation of magmas of primary pacific character. This evidently points out to the comagmatic links of the whole series in the Srednogorie zone.

Irrespective of certain specific features, most plutons show good correlation between the evolution of the homodromous magmatic differentiation and the degree of REE fractionation. This is a criterion for fractionated differentiation of comagmatic series.

2. On the background of a distinct Ce character of ΣTR in all rock varieties of the two petrochemical series, two relatively independent tendencies of geochemical behaviour may be distinguished: the K-subalkaline plutons show higher itrium content while most of the intrusives of Ca-alkaline character are richer in cerium. In this light the noted intra-formational inversion of the REE fractionation in some of the polyfacies plutons is of certain interest. It is established that this change in the course of lanthanoid fractionation is a result

mainly of the quantitative and qualitative composition of the minerals concentrators of REE.

3. Based on the values of Eu, a threefold internal interruption of REE fractionation in the facies development of the plutons is established. This supports the model for the three main stages in the formation of the Middle Alpine intrusions in the Srednogorie zone.

The aplite-pegmatite phase may be divided as an additional stage. This phase shows lower ΣTR (A l e k s i e v, 1969) since at that time independent minerals concentrators of REE are formed — thorite, orthite (B o y a d j i e v — I v a n o v, 1975, 1982).

The dike formation is in principle a relatively new, independent stage of partial magmatic differentiation and lanthanoid fractionation. Typical are considerable variations of the Eu values in individual facies varieties and the recurrent fractionation of ΣTR in the latest lamprophyres.

4. The gabbroids of the first magmatic stage of the composite plutons as well as the lamprophyres of the dike formation mark a definite interruption in the general process of lanthanoid fractionation. Probably this reflects their relatively different genesis and to a certain degree their independent geochemical behaviour.

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