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K-Ar dating of metamorphic rocks from Strandja massif, SE Bulgaria

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Abstract. The stratigraphic ages of the geological bodies that built up the Strandja diabase-phyllitoid complex cover a wide range – from Early Devonian to Middle Jurassic. The mineral paragenesies of the rocks are typical of the greenschist facies and include albite, chlorite, sericite (illite-muscovite), epidote, calcite. The temperature interval of 350-400°C is estimated by the chlorite geothermometer. The b_0 values and the illite crystallinity (IC) values of illite/muscovite indicate high-temperature anchizonal-epizonal metamorphic conditions.

K-Ar isotopic dating of illite/muscovite-rich < 2 µm grain size fractions mainly from representative samples of phyllites and metadiabases leads to the following conlusions. Part of the K-Ar dates of the fractions < 2 µm, < 60 µm, > 60 µm indicate Jurassic (160–170 Ma) low-grade regional greenschist metamorphism. The K-Ar dates 110–150 Ma of the other part of the fractions < 2 µm reflect the rejuvenation effect of the Upper-Cretaceous (80–100 Ma) regional tectonothermal (250-280°C) event related to the subduction of Tethyan oceanic lithosphere.

The primary magmatic and metamorphic ages are Early Paleozoic (probably Early Devonian) based on a poorly defined whole rock Rb/Sr isochron from metadiabases. The K-Ar dates 246 – 309 Ma of some metadiabases confirm their Paleozoic age and indicate at least one Paleozoic low-grade or higher-grade metamorphic event. The ^{207}Pb / ^{206}Pb dates of single zircons from metagranites and metamorphic rocks from the central part of the Strandja massif (Turkey) could be regarded as an indicator for probable regional high-grade Hercynian metamorphism overprinted by Jurassic greenschist metamorphism.

Key words: Strandja Massif, K-Ar dating, metamorphic rocks

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Резюме. Стратиграфската възраст на геоложките тела, изграждащи Странджанския диабаз-филитоиден комплекс покрива широк интервал от ранния девон до средната юра. Минералните парагенези на различните видове скали са типични за зеленошистния фациес и включват, кварц, мусковит, албит, хлорит, калцит. С помоща на геотермометъра на Cathelineau (1988) са получени температури в интервала 350-400˚C. Ако се съди по параметъра b_0 и стойностите на илитната кристалинност (IC) от богати на мусковит фракции < 2 µm на представителни проби от филити и метадиабази, то характерни са били високотемпературни анхизонални-епизонални условия на метаморфизъм.

Извършено е K-Ar изотопно датиране на богати на мусковит < 2 µm зърнести фракции предимно от филити и метадиабази на Странджанския диабаз-филитоиден комплекс. Част от K-Ar датировки на фракциите < 2 µm, < 60 µm и > 60 µm индикират юрски нискостепенен регионален зеленошистен метаморфизъм 160–170 Ma. K-Ar датировки 110–150 Ma на друга част от фракциите < 2 µm отразява подмладяващото въздействие на горнокредното тектонотермично събитие 80–100 Ma, *T* 250–280˚C, кореспондиращо на субдукцията на Тетиската океанска литосфера.

Първичните магматични и метаморфни възрасти са долнопалеозойски (вероятно раннодеонски) и се основават на палеонтоложки данни и на недобре дефинирана Rb-Sr-изохрона от метадиабази. K-Ar датировки на същите метадиабази потвърждават тяхната палеозойска възраст и са индикатор на поне едно метаморфно палеозойско събитие в зеленошистен или по-висок фациес. ^{207}Pb / ^{206}Pb датировки 228–309 Ma на единични циркони от метагранити и метаморфити могат да се разглеждат като индикатори на вероятно високостепенен херцински метаморфизъм с наложен върху него юрски зеленошистен метаморфизъм.

Introduction

The Strandja massif is located in the south easternmost part of the European continent. The term was introduced by Pamir and Baykal (1947) to denote a considerable part of the territory of Bulgaria and Turkey, including Sveti Ilya and Manastir Heights, Sakar Mountain, Dervent Heights and Strandja. Recently, this viewpoint has been adopted by Okay et al. (2001). According to Dimitrov (1956, 1958), this was an area of intensive Jurassic diabase and granite magmatism and meso- to epizonal regional metamorphism. He considered the massif to be an independent tectonic unit – "Strandja-Sakar tectonic zone". Boncev (1946) understood the Strandja massif as a part of a "mobile Balkan space" (referred later to the Eastern Srednogorie), build up of two major structures: the Strandja anticlinorium to the southwest and the Burgas synclinorium to the northeast. Čatalov (1990) assumed that the Strandja massif, renamed to "Strandja zone", consisted of two superposed allochthonous units – lower (East-Thracian nappe) and upper (Zabernovo nappe). The two units were emplaced in their present position as a result of two global tectonic events. The first one was related to the closure of the Paleotethys and the second – to the closure of the Neotethys. The presence of Triassic rock sequences, formed in different structural and facial zones, was pointed out to be a characteristic feature of the massif. Their present superposition was assumed to result from the thrusting events. According to Čatalov (1990), all pre-Cretaceous rocks in the Strandja massif were affected by greenschist metamorphism with a "reversal" of the isogrades.

Metamorphic rocks are widespread in Strandja massif. Many years of detailed lithostratigraphic studies proved pre-Cambrian, Paleozoic, Triassic and Jurassic rock units. The attention was focused on the greenschist metamorphic rocks from a thick diabase-phyllitoid complex. We shall conventionally denote the latter "Strandja diabase-phyllitoid complex", despite its compositional, structural, tectonic and metamorphic afinities to Lower Paleozoic rocks from the Central and Western Balkan. A considerable part of the rocks within this complex are Early Palaeozoic in age, including Early Devonian (Maliakov, Prokop, 1997; Boncheva, Catalov, 1998; Lilov, Maliakov, 2001a, b). Rocks of similar age were reported to the west of the Sakar mountain (Haskovo-Harmanli region) and in the Dervent Heights (Latcheva et al., 1989; Lakova, 1998, and others). All these data shed new light on the problems of the lithological volume, the lithostratigraphic subbdivision, the interrelations between rocks bodies, their chronostratigraphic range, the metamorphic and tectonic events of the metamorphic rocks from the Strandja massif and adjacent areas.

The aim of the present paper is to test the possibilites of K-Ar dating of layered silicates, formed in low-grade metamorphic *P-T* conditions. Based on the avaiable isotopic dates of magmatic and metamorphic rocks from the Strandja massif and data from new studies, an attempt is made to obtain information on the age of the different geological events that affected the Paleozoic and Mesozoic sequences of the massif.

Stratigraphy

About 85% of the Strandja diabase-phyllitoid complex comprises various in composition phyllites, closely interbedded with marbles, quartzites, metaarcoses metasandstones, metasiltstones, products of submarine diabase magmatism, etc. The distribution of the complex and the location of the collected samples is shown in Fig. 1.

The complex overlies diverse basement rocks which are locally preserved in western Strandja and Sveti Ilya Heights. The section starts with metaarkoses and metasandstones topped by black phyllites with metasilstone and marble interbeds (Stravnica Formation, after Čatalov, 1985b, 1990). Upwards follows a thick unit of black phyllites with metasiltstone intercalations (Stoilovo and Zabernovo Formations, after Čatalov, 1985b). Metasiltstones predominate in the lower part, and phyllites – in the upper one. This unit is overlain through a rapid lithological transition by black or green calcophyllites (Gramatikovo Formation, Čatalov, 1985b). The transitional zone between the two units may be observed in many outcrops in Central and Southeastern Strandja, in the preserved overturned limb of the ”Strandja” synschistous fold-nappe (in the sense of Maliakov, 1976). This is the zone where metadiabases and various products of submarine diabase magmatism tend to concentrate. The section terminates with a thick marble association (Malko-Turnovo Formation, Čatalov, 1985b). In general, the described section correlates well with the stratigraphic sequence in Turkish Strandja (Aydin, 1974; Okay et al., 2001).

Tectonic and metamorphic events

The rocks of the Strandja diabase-phyllitoid complex were repeatedly deformed. Various structural elements of deformational origin can be observed. The geometrical analysis of their relationships proves that they were formed as a result of a succession of several phases of deformation. The first and the second one were more important. They produced two generations of penetrative planar and linear structures of regional distribution cleavages S_1 and S_2, lineations L_1 and L_2, fold structures B_1 and B_2, etc. (Fig. 2). They can be distinguished both morphologically and in time and space. After the first phase, all followig deformations were controlled by the surfaces of the first cleavage S_1.

S_1 is the the earliest planar structural element of deformational origin. According to morphological features, this is a typical foliation cleavage. The cleavage planes are marked by the planar orientation of phyllosilicates, actinolite, albite, quartz in rather variable quantitative relationships. Commonly the S_1 planes host synmetamorphic quartz or quartz-carbonate veins of different thickness – from several mm to several tens of cm.

The second regional cleavage S_2 differs by morphology from the first cleavage and varies between typical “fracture“ and “microcrenulation” cleavage, depending on the mechanical behaviour of the deformed rocks (phyllite, metadiabase, metaarcose, metasiltstone, marble, etc.). For this reason the distance between cleavage planes varies between 0.5-1 mm and 1-2 cm (Fig. 2).

The second deformation phase affects all pre-Upper Cretaceous rocks, e.g. the rocks of the Strandja diabase-phyllitoid complex, the Triassic and Jurassic sequences. It was likewise accompanied by a regional greenschist metamorphism. The metamorphic recrystallization affected all rocks but to a different extent. According to microscopic studies, its effect is best expressed around the cleavage planes S_2. The interal parts of the microlithons reveal a typical arcuate pattern of the post-tectonic rock-forming minerals that fossilize the new, deformed position of S_1. The recrystallization was syntectonic and dominantly post-tectonic with the appearance of a second generation of minerals that are typical of greenschist facies metamorphism. In priciple, it caused a progressive greenschist metamorphism of the Triassic and Jurassic sediments whithout essentially affecting the Stranja diabase-phyllitoid complex that was already metamorphosed in greenschist facies.

Petrographic notes

The object of this study are mainly phyllites and metamorphosed in greenschist facies diabases. Macroscopically, phyllites are black

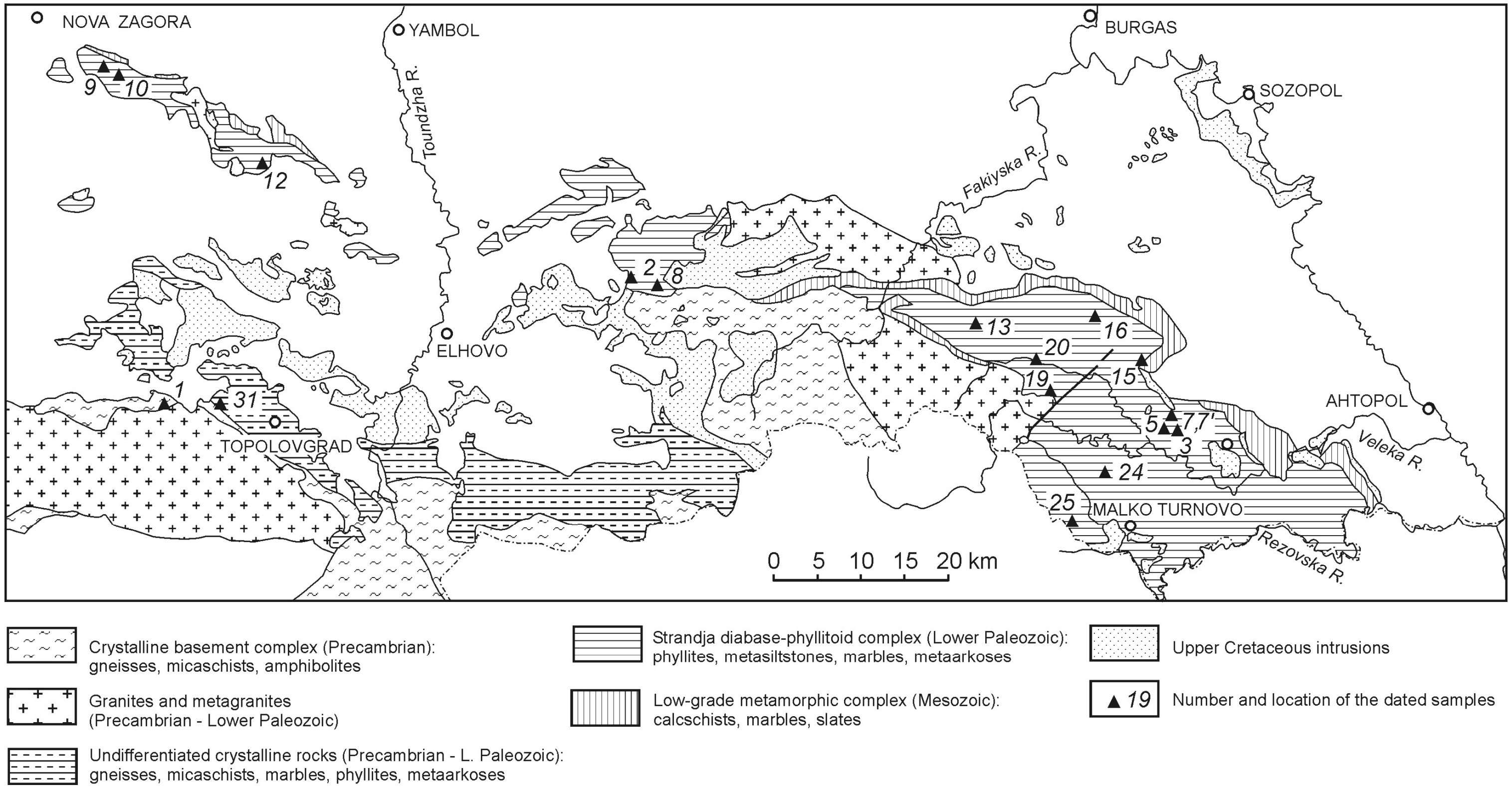

Fig. 1. Schematic geologocal map ot the Southeast Bulgaria with number and location of the dated samples
Фиг. 1. Схематична геоложка карта на ЮИ България с номер и място на датираните образци

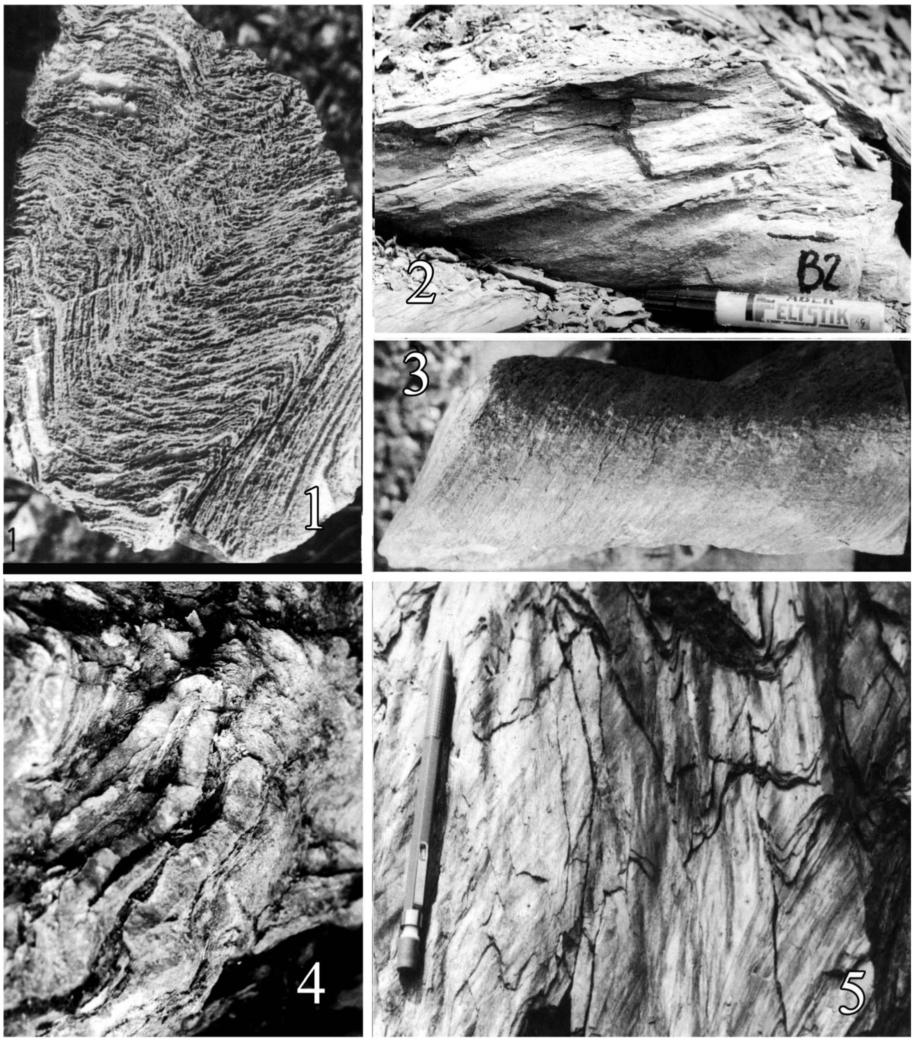

Fig. 2. Two stages of tectonic deformation and metamorphic re-crystallization of rocks from Strandja diabase-phyllitoid complex. The schistose cleavage S_1 is in all cases deformed and folded into narrow to isoclinal B_2 folds, which deform the older intersection lineation L_1. Cm-thick synmetamorphic quartz-carbonate veins, parallel to S_1, are folded together with S_1. The axial plane of B_2 folds is fracture cleavage S_2. 1) and 2) Calc-schists from Dimov Pazlak locality, Malko Turnovo area and from the confluence of Stravnitsa into Veleka River; 3) marbles at Grudska Chuka locality, Malko Turnovo area; 4) metasiltstones in the valley of Stravnitsa River, west of Valkancha locality, and 5) phyllites with relictic bedding in Dokuzaka locality, south of Stoilovo Village

Фиг. 2. Два етапа на тектонски деформации и метаморфни прекристализации на скалите от Странджанския диабаз-филитоиден комплекс. Шистозният кливаж S_1 е деформиран и огънат в тесни изоклинални B_2–гънки, които усукват ранната линейност на пресичане L_1. Сантиметрови синметаморфни кварц-карбонатни жили, паралелни на S_1 са огънати заедно с нея. Осова плоскост на B_2–гънките е кливажът S_2. 1) и 2) Калкошисти от местн. Димов пазлак, Малкотърновско и от разкритие при устието на р. Сръвница в р. Велека; 3) мрамори от местн. Грыцка чука, Малко-търновско; 4) метаалевролити от долината на р. Стръвница, западно от местн. Вълканча; 5) филити с реликтова слоестост от местн. Докузака, южно от с. Стоилово or grey rocks with fine-schistose structure. Under the microscope they show a relatively constant mineral paragenesis with main rock-forming minerals albite, white mica, chlorite, epidote, actinolite, quartz and calcite. X-ray diffractometry (XRD) investigations of the phyllosilicates prove that the chlorite is everywhere represented by by the IIb-polymorphic modifications and the white mica by the polymorphic modifications $2M_1$, considered by Stefanov (2000) to be illite/muscovite. In the calcite-containing phyllites, the quantity of clinozoisite or epidote is nearly constant. The quantity and distribution of coalified substance or finely dispersed pyrite in the form of microscopic globules is rather irregular. The black and grey-black colour of the phyllites is namely due to them. Accessory minerals are zircon, monazite, tourmaline, apatite, ilmenite, titanite, the latter commonly altered to leucoxene. The texture of the rocks is mostly fine-grained microlepido-blastic. In phyllites, in which relictic layered structure is preserved (2–3 mm thick metasiltstone layers), part of the muscovite is terrigenous, between 70–150 µm in size.

The metadiabases are green to dark-green rocks with rough schistose structure. Main rock-forming minerals are albite and chlorite, minor – epidote, calcite, white mica (sericite), accessory apatite and titanite. Albite forms ephedral individuals with cores filled by epidote grains. The angular cavities between albite grains are occupied by single crystals or aggregates of brown-green chlorite, optically positive. Part of the chlorite is replaced by post-tectonic muscovite. Around the contacts with the host rocks, the diabases are altered into green rocks built up of a microgranular ground mass of albite, chlorite, epidote, sericite, quartz and ore substance - most commonly pyrite. The primary ophitic texture is preserved only in the internal parts of the diabase bodies into microlithons 2–5 mm to 1 cm thick (Lilov, Maliakov, 2000). Near the schistose surfaces, the lamelae of the albitic grains are bended and a large part of them are fragmented. Typical of the rocks is peripheral granulation of the main minerals, unclear diffuse contacts of the brown-greenish chlorite, appearance of post-tectonic, later optically negative chlorite, post-tectonic sericite, albite without inclusions and quartz with mosaic structute.

By applying of chloritic geothermometer of Cathelineau (1985, 1989) samples collected around the Zvezdec Village and to the south of Brashlyan Village, were used to study the temperature conditions of the first and second regional greenschist metamorphism. The obtained temperatures are between 375 and 410^{o}C. Data on the second metamorphic re-crystallization were obtained by means of the calcite-dolomite geotermometer (Bikle, Powell, 1977; Powell et al., 1984). Rock samples from the marblized limestones of Lipachka formation were studied. The results are a little lower than those obtained by the chloritic geothermometer and scatter between 365 and 385^{o}C.

K-Ar dating of regional tectonothermal events in Southern Bulgaria

The retention of radiogenic isotopes in rock-forming minerals can be relaxed by superimposed chemical and thermal processes during their early or late post-crystallization existence. The effect of the thermal influence on the argon loss from different minerals has been studied for a long time. About 35 years ago, this effect was investigated in natural conditions by Hart (1964) and in the laboratory by Fechting and Kalbitzer (1966). Calculating geochronological information using direct mathematical expressions for the diffusion of radiogenic isotopes is rather difficult because of the necessity of complicated parameters to be measured.

On the basis of theoretical argumentation (Dodson, 1979) for diffusion of radiogenic isotopes in minerals and rocks, and experimental data on the thermal influence on syngenetic K-contained minerals, a thermo-chronic model for regional tectonothermal events in Southern Bulgaria was proposed by

Lilov (1990). Biotite was used as main mineral geothermochronometer. Monomineral biotite fractions mainly from intrusive magmatic bodies older than the age of the studied tectonothermal event were dated. One regional late Cretaceous (80–100 Ma) tectonothermal (250–280 ^{o}C) event in Central and Eastern Srednogorie and Strandja was registered by K-Ar dates (110–140 Ma) of biotites from Paleozoic magmatic and metamorphic rocks (Lilov, 1990). According to the thermochronic model, the pre-Cretaceous magmatic and metamorphic complexes were subjected to one regional tectonothermal effect during the Late Cretaceous. This effect can be regarded as expression of global tectonothermal processes which, in geodynamic aspect, were related to subduction of the Vardar-Intra-Pontide ocean under the Rhodopes (Boccalleti et al., 1974, 1978).

According to generalized K-Ar and Rb-Sr geochronological information and the thermochronic model (Lilov, 1990), the magmatic and metamorphic complexes in the Rhodope massif were subjected to one regional tectonothermal effect during the Paleogene (30–40 Ma). This influence, with temperatures more than 350^{o}C, was controlled by continental collision during the final closure of the Tethys ocean and blocking of the subduction processes (Innocenti et al., 1984).

Zircon dates of metagranites and metamorphites from Strandja Massif

Single zirkons from the metagranite plutons of Kirklareli, Kula and Üsüp and from the basement metamorphic rocks in Central Southwestern Strandja (Okay et al., 2001) were dated by the method of stepwise Pb evaporation technique. The ^{207}Pb/^{206}Pb dates of the investigated zircons are between 228–309 Ma. The authors assume that magmatic crystallization ages of the Kirklareli and Kula pluton are 271 Ma and those of the Üsüp pluton – 309 Ma. Two ages were obtained for zircons in gneisses (266 and 239 Ma) and migmatites (285 and 228 Ma). It was assumed that the age 285 Ma of the migmatic zircon indicates high-grade metamorphism and partial melting during the earliest Permian. However, it is not clear what was the effect of this metamorphism over the zircons from the metagranites and the gneiss from basement. Okay et al. (2001) made an essential mistake about the blocking (isotopic closure) temperature for U and Pb, which in Kober (1986) is 900 K (Kelvin) but not 900°C. The blocking temperature for U and Pb is ~627°C. Close to that temperature of 627°C, partial loss of radiogenic Pb isotopes from the zircons might be possible during the Jurassic (160–170 Ma, this paper) regional greenschist facies metamorphism. The rocks of the crystalline basement of Western Strandja and Dervent Heights on the territory of Bulgaria show distinct traces of brittle-ductile deformations in greenschist facies 400–450°C and P < 8 kbar conditions. Without going into details, it must be noted that there are similar data for Turkish Strandja (Caglayan et al., 1990).

Because of the erroneously defined closure temperature (> 900°C), the Triassic isotopic ages (238 and 239 Ma) of zirkons from the gneiss and the migmatite are difficult to interpret (Okay et al., 2001). The authors consider that a loss of radiogenic Pb isotopes from zircon is not possible at temperature < 900°C. They can not explain when the zircons of Triassic (239 and 228 Ma) age in the basement rocks were formed, since Triassic and Lower-Middle Jurassic sedimentary sequences cover this basement (Čatalov, 1990).

In fact, the published dates of zircons convincingly prove Hercynian or older age of the metagranites, gneisses and migmatites that form the basement of Strandja Massif. Undoubtedly, obtaining precise U-Pb concordia or discordia of zircons from metagranites will allow a more accurate age determination of the magmatic crystallisation and the age of the last metamorphism. We are waiting with interest a competent discussion of the ^{207}Pb/^{206}Pb dates of the zircons in question (Okay et al., 2001) from experts in the interpretation of Rb/Sr and K/Ar geochronologic data (Skenderov, Skenderova, 1995; Ivanov et al., 2001).

K-Ar dating of metamorphic illite/muscovite

The use of illites for K-Ar dating of stratigraphic events is limited. The main obstacle is the presence of detritial grains which preserve partly their radiogenic argon during transport and redeposition and which cannot be separated from authogenic minerals during laboratory treatment of the samples. This has been yet shown since the first K-Ar studies on clay minerals (Hurley et al., 1961; Bailey et al., 1962; Hower et al., 1963). On the other hand, their importance for dating of very low-grade metamorphic events was demonstrated by Hunziker et al. (1986). The disturbing effect of the inherited K-white mica can be eliminated by using the fine-grain size fractions of phyllites and slates (Frank, Stetter, 1979; Clauer, Kroner, 1979; Hunziker et al., 1986; Reuter, 1987; Balogh et al., 1990; Arkai et al., 1995).

The possibilities for isotopic dating of metamorphic events have been greatly improved by the introduction of the concept of blocking temperature (Dodson, 1973) and its determination for the main rock-forming minerals (Wagner et al., 1977; Harrison, McDougall, 1982, and others).

Blocking temperature of 260±30°C for illites of < 2 μm has been estimated by Hunziker et al. (1986). Since re-crystallization may occur at lower temperature, the interpretation of K-Ar data requires a versatile study of mineral fractions (e.g., Hunziker et al., 1986).

During diagenesis, corresponding to very low-grade metamorphic evolution of pellitic rocks, K-Ar ages of illite/muscovite are continuously diminishing from the age of detritus (or from the age of sedimentation) to the age of metamorphic events. Mineralogicaly, this process is characterized by the change from illite/smectite mixed layer clay mineral through illite (fine grained dioctahedral mica type clay mineral with K-deficiency and incorporated smectite layers and H_2O) up to metamorphic muscovite. This process is characterized by gradual decrease of H_2O and increase of the K-content, by increase of the $2M_1/1Md$ ratio which is reflected in the illite crystallinity parameters.

In case of anchizonal and epizonal (200–400°C) rocks, reliable data can be obtained from K-Ar dating of illite/muscovite-rich fractions with grain size < 2 μm. Due to different reasons, separation of monomineral fractions cannot be obtained in general. In practice, this fractions are mixtures of dominanting illite/muscovite with smaller amounts of chlorite, quartz, and other minerals (see also Hunziker et al., 1986).

The temperature range of very low-grade metamorphism overlaps the closure range of the K-Ar system in illite/muscovite. Consequently, special attention should be paid to the distinction between "metamorphic" ages, which refer to the time of crystallization at or near the metamorphic climax and the "cooling" ages, which indicate the time when the K-Ar system became closed.

Although the K-Ar method has been widely used for determination of metamorphic ages of low-temperature rocks in the last nearly thirty years, this is the first attempt to apply this method to solve age problems in such regions in Bulgaria.

Methods

K-Ar isoptope measurements were preceded by microscopic and X-ray diffractometric (XRD) investigations. Illite/muscovite-rich samples from various tectonic and stratigraphic localities were studied. Approximately 2 kg of material from each sample was ground. The grain size fractions < 2 μm, 2-60 μm, > 60μm were separated using sedimentation methods. These fractions are rich in illite/muscovite, but pure white mica concentrates could not be obtained from them. As shown by X-ray diffraction, the low potassium contents of some of the samples are due to considerable amounts of chlorite and minor quartz in them.

X-ray diffraction studies (joint studies with D. Stefanov) of fractions below 2 μm were done by an automated diffractometer D-500 Siemens at goniometer speed 0.5°min^{-1} in the Geological Institute, Bulgarian Academy of Sciences. The data on Kubler's illite crystallinity index (IC) were calibrated on the basis of samples kindly presented by Dr. Arkai, Hungary, calibrated by him from original standards provided by the author of the method Prof. B. Kubler, Switzerland.

To denote different metamorphic stages in the post-depositional evolution of the rocks, Kubler (1968) and other authors after him defined the terms of diagenetic zone, anchizone and epizone. This subdivision is based on the illite crystallinity index (IC) of Kubler. For the anchizone Kubler's Neuchatel limits 0.42/0.25°Δ2θ at $2°min^{-1}$, are equivalent to Kirch's 0.38-0.37/0.21°Δ2θ at 0.5 or 0.6° min^{-1}.

K-Ar dating was performed with instruments constructed in the Institute of Nuclear Research, Hungarian Academy of Sciences. Samples were degassed by high frequency induction heating. Argon was cleaned using the usual method of applying zeolite, cold traps and furnaces with a Ti sponge and CuO. ^{38}Ar was introduced with a gas pipette. For Ar isotopic ratio measurements mass spectrometer of 150 mm radius and 90° deflection was used in the static mode. Before K-determination, the samples were digested by mixing of HF + H_2SO_4 + $HClO_4$ and dissolved there after in HCl. Sodium buffer and Li internal standards were added and K-content was measured with a flame emission photometer. The inter-laboratory standards Asia 1/65 and GL-0 were used for controlling and calibration of Ar and K determination. Errors of K-Ar ages (2σ) were calculated assuming a 3% error for the determination of K. Constructions and parameters of instruments, as well as the applied methods, have been described in detail by Balogh (1985). The results obtained on inter-laboratory standards have been also published. (Inter-laboratory standards for dating purposes, 1982). Atomic constants suggested by Steiger and Jäger (1977) were used for calculating the age. The geological interpretation of the isotopic age data is based on the time-scale of Harland et al. (1982).

Results

X-ray diffractometric analyses

Semi-quantitative mineral composition and illite crystallinity (IC) of 11 samples and b_0 parameter of 6 samples (Tabl. 1) were determined by the X-ray powder diffractometric (XRD) method. According to XRD data, the mineral composition of the studied metapellites includes fine dispersed minerals that dominate mineral fractions below 2 μm. Major rock-forming minerals are dioctaherdal micas and thrioctahedral chlorites. The mica contains variable amount of K and Na cations in the inter-layers and Fe and Mg cations in octahedral and tetrahedral position. Because of this reason, following Stefanov (2000), white micas here and in the following text are denoted as illite/muscovite. Second in significance is chlorite while the rest of phyllosilicates (paragonite and pyrophyllite) are in small amounts. Other minerals are quartz, feldspar and hematite.

K-Ar dates measured on illite/muscovites are listed in Table 1 together with mineral composition, illite crystallinity data and b_0 parameter. The IC and b_0 data (Table 1) are statistical ones and for this reason no direct conclusion can be drawn for the metamorphic grade and pressure conditions. The conclusions made below are based on the data from Table 1 in Stefanov (2000) and unpublished data for b_0.

Illite crystallinity

The IC values and the b_0 parameter are shown in Table 1. The Kubler's illite crystallinity index (IC) varies in the range of 0.19 to 0.26°Δ2θ for metasediments and metavolcanics of different geological age from different regions. The IC values of all samples are in most cases close to the boundary between the anchi- and epizone. Only the Jurassic rocks are in the anchizone, while the Triassic and Paleozoic ones fall within the epizone. The generalized results are as follows (Table 1):

1. The typical anchizone is demonstrated by the Jurassic metasediments from Strandja South (IC = 0.26°) and the Sveti Ilya Heights

Table 1. *K-Ar isotopic compositions, apparent ages, mineral composition and illite crystallinity data of the metamorphic rocks from the Strandja massif, SE Bulgaria*

Таблица 1. *K-Ar изотопни състави, привидни възрасти, минерален състав и данни за илитната кристалинност на метаморфни скали от Странджанския масив, ЮИ България*

Sample	Locality	Formation	Rock	Fraction	K%	^{40}Ar(rad)	^{40}Ar(rad)	Age Ma	IC	*bo* Å
				Sveti-Ilya Hights						
1	Orlov Dol	Jaltical	gneiss	0.12-0.06	6.69	3.48	83.1	129.3±4.9		
8	St. Karadjovo	Sokolska	phyllite	<0.002	2.104	0.744	49.2	88.7±3.8	0.19	9.012
9	ts. Karakay	Kavashka	etavolcanit	>0.063	0.434	0.269	60.9	152.8±6.0	0.2	9.007
10	ts. Karakay	Kavashka	etavolcanit	<0.002	4.29	2.036	91.1	118.0±4.5	0.21	9.007
12	Botevo	Zvezdecka	phyllite	<0.002	2.85	1.423	61.5	128±25.1	0.24	8.982
				Strandja Mountain						
13	Bliznak	Zvezdecka	phyllite	<0.002	3.36	2.115	82.5	155.1±5.9	0.26	8.987
15	Kalovo	Kazanska	phyllite	<0.002	3.1	1.526	58.2	122.4±5.0	0.26	8.989
16	Byala voda	Lipacka	phyllite	<0.002	5.23	2.967	85.6	140.3±5.3	0.22	
19	Zvezdec	Gramatikovo	phyllite	>0.063	1.27	0.854	56.7	165.2±6.7	0.19	
20	Mladejka River	Zaberska	phyllite	>0.063	2.23	1.574	89.9	173.0±6.5	0.19	
24	Dligurya	Stoilovo	phyllite	>0.063	2.66	1.86	85.4	171.5±6.5	0.2	
24’	Dligurya	Stoilovo	phyllite	<0.002	4.03	2.476	73	151.5±5.9	0.2	
25	Ay-dere River	Zvezdecka?	phyllite	<0.002	4.45	2.272		126.8±5.0		
3’(26)	borehole 170	Gramatikovo	calcophyllite	<0.002	4.07	2.461	75.9	143.6±5.9		
5’(27)	borehole 154	Zaberska	metadiabase	0.002-0.06	0.234		50.5	203.1±8.3		
7’(28)	borehole 156	Zaberska	metadiabase	<0.002	3	1.589	70.4	131.4±5.4		
7’’(28)	borehole 156	Zaberska	metadiabase	>0.063	0.173		61.7	237.0±9.7		

(IC = 0.26°). The Jurassic metapelites from Strandja North (IC = 0.23°) may be referred to the high-temperature part of the anchizone, close to the boundary with the epizone. The b_0 values of the Jurassic illite/muskovites from Strandja are uniform and close to b_0 = 8.998 Å. This b_0 value is characteristic for the rocks formed at low pressure.

2. The Paleozoic metapelites from Sveti Ilya Heights (IC = 0.19°) and the Triassic metapelites from Strandja (IC = 0.18°) and Sveti Ilya Heights (IC = 0.18°) fall within the epizone. The predominant *P/T* conditions for the Paleozoic rocks were characterized by temperatures 350–400°C, locally reaching the biotite isograd (400–450°C), and pressure 3 kbar, b_0 = 9.007 Å. The b_0 values of the Triassic metapelites vary from 9.014 to 9.037 Å or in general b_0 = 9.029 Å for the Triassic metapelites. These rocks belong to the facial series of medial pressures (Guidotti, Sassi, 1986).

K-Ar dating of illite/muscovite of the metamorphic rocks from Strandja massif

The K-Ar dates (Table 1) of illite/muscovite-rich < 2 μm, 2-60 μm and > 60 μm grain size fractions from Paleozoic, Triassic and Jurassic metamorphic rocks in Sveti Ilya Heights and Strandja are in the range of 118–173 Ma. Only sample 7 contains mainly chlorite and its date is 88.7 Ma. With the exception of sample 9 (metavolcanite, fraction > 60 μm), the dates of the illite/muscovite fractions from Sveti Ilya Heights and from northern part of Bulgarian Strandja are younger (89.1–128 Ma).

The illite/muscovite fractions both in phyllites and metadiabases from Strandja yield older dates (127–173 Ma). The oldest K-Ar dates of fractions < 2 μm, 2-60 μm and > 60 μm are in the range of 155–173 Ma. These data show, that fractions > 60 μm contain insignificant amounts of detrial muscovite as compared to the newly-formed metamorphic $2M_1$ illite/muscovite.

There is a second possibility and namely, that the temperature (400°C) of the Jurassic regional greenschist metamorphism was higher than the blocking (350°C) temperature of detrial muscovite from the Paleozoic, Triassic and Jurassic phyllites. In this tectonothermal environment, an almost total loss of radiogenic Ar from the detrital muscovites took place and the K-Ar chronometer in them started timing from zero before 160–170 Ma.

The K-Ar dates (between 160–170 Ma) of the illite/muscovites fix the age of the mid-Jurassic low-grade metamorphic event. According to IC and b_0 data, the pre-Jurassic rock formations suffered Jurassic (160–170 Ma), regional epizonal greenschist facies metamorphism at low temperatures (between 350–450°C) and low to medium pressure (3–5 kbar).

There are no significant differences between the < 2 μm illite/muscovite K-Ar data from different rock types. There is a well expressed tendency to a more significant rejuvenation (89–128 Ma) of these dates when nearing the Srednogorie Upper Cretaceous magmatic arc. The dates in question are scattered between the ages of the Jurassic and the Upper Cretaceous event. Thus, the larger dispersion of these dates can be interpreted by partial resetting of the K-Ar system of Jurassic illite/muscovite with blocking temperature 260±30 ˚C during the Upper Cretaceous (80–100 Ma) regional tectonothermal (250–280°C) event (Lilov, 1990) manifested in the Central and Eastern Srednogorie. This event can be related to the northward-subducting Tethyan oceanic lithosphere under the Rhodopes and Strandja (Boccaletti et al., 1974, 1978) and isotherm upraisal within the continental crust of Strandja massif.

Conclusions

The primary magmatic and metamorphic ages of the rocks from Strandja diabase-phyllitoid complex are Early Paleozoic. Early Devonian age (~386 Ma) was obtained from a poorly defined whole rock Rb/Sr isochron of metadiabases from the volcano-sedimentary complex in Central Strandja (Lilov, Maliakov, 2001a). K-Ar dates (246–305 Ma) of the Devonian metadiabases confirm their Paleozoic age (Lilov, Maliakov, 2001b) and show that the pre-Triassic rock formations were affected by at least one Paleozoic metamorphic event.

The $^{207}Pb/^{206}Pb$ dates (228–309 Ma) of single zircons from metagranites and metamorphic rocks (Okay et al. 2001) could be regarded as indicators of their Hercynian or older age. The zircons suffered partial loss of radiogenic Pb under the influence of the Jurassic (160–170 Ma) regional greenschist facies metamorphism.

A regional mid-Jurassic (160–170 Ma) low-grade greenschist metamorphism with temperature 350~450°C and pressure 3–5 kbar is indicated by K-Ar dating of illite/muscovites from phyllites and metadiabases. The pre-Triassic rocks suffered more than one low-grade greenschist metamorphism but K-Ar dating of illite/muscovites records only the last metamorphism.

The regional Late Cretaceous (80–100 Ma) tectonothermal (250–280°C) event (Lilov, 1990) can be related to the northward-subduction of Tethyan oceanic lithosphere under the Rhodopes (Boccaletti et al., 1974, 1978). This event is recorded by the K-Ar dates (110-150 Ma) of biotites from pre-Cretaceous magmatic and metamorphic rocks (Lilov, 1990) and rejuvenated K-Ar dates (100–150 Ma) of Jurassic (160–170 Ma) illite/muscovites from pre-Jurassic phyllites and metadiabases.

The regional Paleogene (30–40 Ma) tectonothermal event within the Rhodope massif (Lilov, 1990) did not affect the rock sequences in Strandja. So far, no Paleogene isotopic datings of magmatic and metamorphic rocks from Bulgarian Strandja have been obtained.

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