GEOLOGICA CARPATHICA, DECEMBER 2010, 61, 6, 469–481 doi: 10.2478/v10096-010-0029-9

Eoalpine (Cretaceous) very low- to low-grade metamorphism recorded on the illite-muscovite-rich fraction of metasediments from South Tisia (eastern Mt Papuk, Croatia)

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(Manuscript received February 11, 2010; accepted in revised form October 13, 2010)

Abstract: Eoalpine very low- to low-grade metamorphism related to Cretaceous orogenesis has been investigated in the Slavonian Mts, Croatia. Samples belonging to the Psunj metamorphic complex (PMC), the Radlovac metamorphic complex (RMC) and Permian-Triassic and Triassic sedimentary sequences (PTSS) were studied. The Kübler and Árkai indices of all the analysed samples indicate high-anchizonal to epizonal metamorphism. The degree of Eoalpine metamorphism tends to be constant in all samples implying that the different complexes passed through and recorded the same event. Measurements of illite-white K-mica b_0-parameter of the RMC samples imply transitional low- to medium-pressure character of the metamorphism. These data together with K-Ar ages (~100–80 Ma) measured on illite-white K-mica rich <2 μm grain-size fractions point to Late Cretaceous very low- to low-grade regional metamorphism presumably related to the main nappe-forming compressional events in the Pannonian Basin and the Carpathians. The P-T-t (pressure-temperature-time) evolution of the studied area is in good agreement with similar scenarios in the surrounding areas of Tisia, but also in Eastern Alps, Carpathians and Pannonian Basin (ALCAPA).

Key words: Eoalpine metamorphism, South Tisia, Radlovac metamorphic complex, thermobarometry, geochronology, Kübler index, Árkai index, K-Ar dating.

Introduction

The Tisia Unit in the sense of Csontos et al. (1992), which is equivalent to the Tisia Megaunit of Szederkényi (1996), represents a large lithosphere block with a complex internal structure. It has traditionally been regarded as one of the most stable parts of the Pannonian Basin basement which practically escaped Alpine metamorphism. Over the last few decades several researches (Árkai et al. 1998, 2000; Árkai 2001 and references therein) pointed out that this is not completely true, showing that large areas of the Pannonian Basin basement were in fact metamorphosed or overprinted as a result of the Eoalpine (Cretaceous) metamorphism.

The Tisia Unit is made up of Variscan igneous and metamorphic complexes and post-Variscan overstep sequences. Typical Tisia rocks are widespread and can be found in the South Transdanubian ranges (Mecsek, Villány Hills), in the Apuseni Mountains (Bihor, Pădurea Craiuli, Codru-Moma, Higniş) and in the Slavonian Mountains (Psunj, Papuk, Krndija), where some of the best outcrops of the South Tisia igneous and metamorphic rocks occur in Mt Papuk (e.g. Pamić & Jurković 2002; Pamić et al. 2002). These rocks, which can provide detailed insight into the geological history of the Tisia Unit form polymetamorphic complexes (Pamić & Lanphere 1991). The complexes of the Slavonian Mts are subdivided on the basis of structural and lithostratigraphic studies by Jamičić (1983, 1988). He distinguished three metamorphic complexes with distinct metamorphic evolution paths; namely the Psunj metamorphic complex, the Papuk metamorphic complex and the Radlovac metamorphic complex.

The present study focuses mostly on the Radlovac metamorphic complex (RMC) which consists of very low- to low-grade metamorphic sequences locally intruded by metabasic rocks (Pamić & Jamičić 1986). According to Jamičić & Brkić (1987) and Jamičić (1988) the RMC occupies the highest structural position of all the Variscan complexes in the area and is uncomformably overlain by Permian-Triassic and Triassic sedimentary sequences (PTSS). The RMC represents the original sedimentary cover over the PMC (Jamičić 1988). Although some previous studies indicated a certain P-T evolution of this area (Jamičić 1983, 1988; Slovenec 1986; Pamić & Lanphere 1991; Jerinić et al. 1994; Judik et al. 2002; Biševac et al. 2009), no research has been carried out so far in order to constrain the spatial, temporal and metamorphic conditions of a possible Eoalpine metamorphism in this area. In this paper a more detailed study concerning the degree and age of metamorphic evolution of this area in the specific geological timeframe is presented and correlated with similar investigations carried out in other parts of the Tisia Unit and surrounding areas.

The main aim of the present paper is to provide new data on the nature and regional distribution of the Eoalpine metamorphism that affected the Variscan and post-Variscan formations of the southern part of the Tisia Unit in the Slavonian Mts. For this purpose, rocks belonging to the RMC as well as those representing the PTSS and parts of PMC sampled along the Kutjevo transect (Balen et al. 2006) were studied by the illite "crystallinity" (Kübler index), chlorite "crystallinity" (Árkai index) and vitrinite reflectance methods in order to determine the temperature conditions, while the b_0-parameter of K-white mica was used in order to estimate the pressure conditions of metamorphism. The K-Ar age dating method was used to determine the age of metamorphism and metamorphic overprint of the studied rocks.

The new P-T and age data presented in this paper form a solid base for further research and reconstruction of the tectono-metamorphic history of the wider area and its correlation with similar metamorphic complexes in Central and Eastern Europe.

Geological settings

The Slavonian Mts, situated in Slavonia, the north-eastern region of Croatia (Fig. 1), are characterized by numerous outcrops of igneous and metamorphic rocks that form the Tisia Unit. Tisia represents a continental fragment broken off from the southern rim of the Eurasian plate (i.e. the southern margin of Variscan Europe) during the Alpine evolution of Tethys (Géczy 1973). After complex drifting and multiple rotation events, mostly during Mesozoic and Cenozoic times, the Tisia reached its present tectonic position (Csontos 1995; Haas & Péró 2004) (Fig. 1). The Tisia Unit is made up of three nappe systems (Mecsek, Villány-Bihor and Békés-Codru) comprising the Variscan igneous and metamorphic basement and post-Variscan overstep sequences (Haas & Péró 2004; Csontos & Vörös 2004; Schmid et al. 2008). According to the interpretation of Schmid et al. (2008) the Slavonian Mts represent an integral part of the Villány-Bihor nappe system. The Variscan igneous and metamorphic complexes of south Tisia, which crop out in the Slavonia region, constitute a part of the metamorphic belt characterized by granitoid rocks accompanied by migmatites (Pamić & Lanphere 1991). These rocks can be well correlated with similar rocks in the Mecsek Mts (south Hungary) (Buda

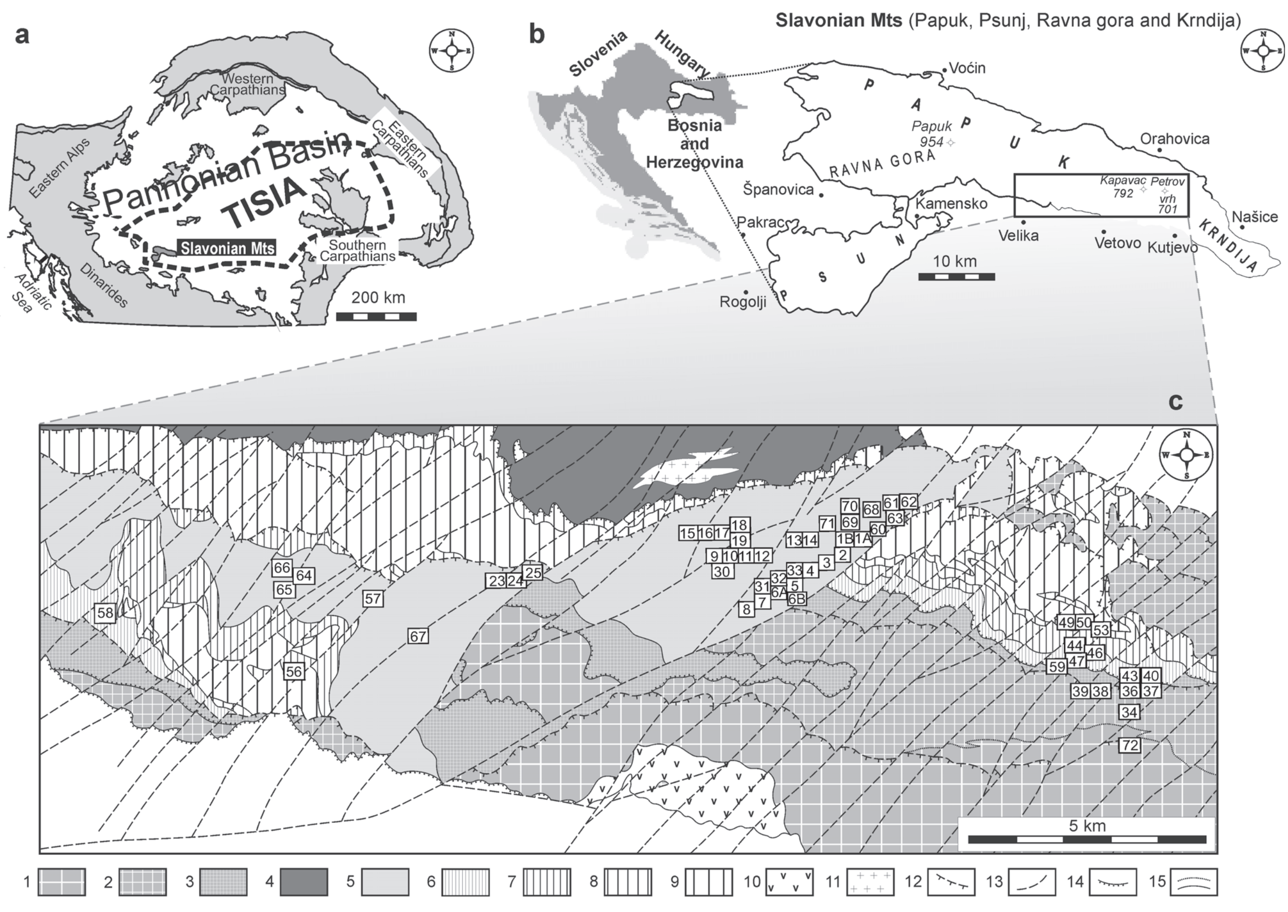

Fig. 1. a — Tectonic setting of the Tisia Unit within the Alpine-Carpathian-Dinaric framework with the position of the Slavonian Mts. **b** — Sketch map of the Slavonian Mts (Papuk, Psunj, Ravna gore and Krndija) with approximate position of the studied area (marked by the black box). **c** — Simplified geological map of the investigated area showing the position of the samples within the complexes as defined by Jamičić (1988). **1** — garnet-staurolite gneisses with lenses of granitoids, amphibolites and amphibolite schists; **2** — chlorite-sericite schists; **3** — metagreywackes and chloritoid schists; **4** — migmatites; **5** — slates, phyllites, quartzites, metasandstones and metaconglomerates; **6** — phyllitic metaconglomerates; **7** — quartz metasandstones; **8** — metasandstones; **9** — dolomites and dolomitic limestones; **10** — gabbro; **11** — granitoid; **12** — reverse fault (covered or assumed); **13** — normal fault (covered or assumed); **14** — erosive or tectono-erosive boundary; **15** — normal boundary (established, covered). Remark: 1 and 2 belong to Psunj metamorphic complex; 4 represents part of the Papuk metamorphic complex; 3 and 5 represents the Radlovac metamorphic complex; 6, 7, 8 and 9 belong to the Permian-Triassic sedimentary sequence.

1981; Hass & Péró 2004), the Western Carpathians (Hovorka & Petrík 1992) and in other European Variscan terrains, such as the Bohemian Massif (Liew et al. 1989).

Jamičić (1983, 1988) distinguished three tectono-metamorphic complexes in the Slavonian Mts, characterized by several phases of deformation and metamorphism (Fig. 1). The *Psunj metamorphic complex (PMC)* is assumed to be formed by a metamorphic event during the Baikalian orogeny showing a strong retrogressive overprint as a result of the Caledonian orogeny. According to Pamić et al. (2002) the rocks belonging to the PMC together with Papuk metamorphic complex, as defined by Jamičić (1988), represent a Barrovian metamorphic sequence, characterized by zonal distribution of index minerals, ranging from greenshists to amphibolite facies conditions. Greenschist facies metamorphic sequences are composed of metapelites, chlorite schists and micaschists, while amphibolite facies sequences comprise paragneisses, garnetiferous micaschists, amphibolites, metagabbros and marbles, locally intruded by discordant granodiorites and plagiogranites (i.e. I-type granites according to Pamić 1986; Pamić & Lanphere 1991). Although geochronological data point to the main metamorphic phase closely associated to the Variscan orogeny (Pamić et al. 1988, 1996; Pamić 1998), Balen et al. (2006) proved the existence of a pre-Variscan metamorphic event. Recent studies (Biševac et al. 2009) showed that PMC also experienced post-Triassic very low- to low-grade metamorphic overprint. The *Radlovac metamorphic complex (RMC)* consists of very low- to low-grade metamorphic sequences largely composed of slates, metagreywackes, metaconglomerates and subordinate phyllites. According to Jamičić (1988) it was metamorphosed during the late stages of the Variscan orogeny. The lower and middle parts of the RMC are intruded by metabasic rocks (Pamić & Jamičić 1986). The age of sedimentation, but also the age of metamorphism of the RMC, arouses the curiosity of many researchers. According to the field relations the RMC unconformably overlies the prograde metamorphic sequences (Jamičić 1983, 1988) and contains a Westphalian microflora (Brkić et al. 1974) which documents a Pennsylvanian age of the protolith. Jerinić et al. (1994) state that the protolith rocks are Silurian, Devonian to (?) Mississippian. K-Ar dating of clinopyroxene monomineralic concentrate from ophitic metagabbro, which intruded the complex (Pamić & Jamičić 1986), gave ages of 416.0 ± 9.0 and 318.6 ± 12.2 Ma (Pamić et al. 1988; Pamić & Lanphere 1991). K-Ar age determinations on two slates from different complexes yielded K-Ar whole rock ages of 203.9 ± 6.9 Ma (slate belonging to the RMC) and 100.6 ± 3.5 Ma (slate belonging to the PMC), which, according to Pamić et al. (1988) apparently represent partially or completely reset ages, due to the subsequent heating. The available data are not sufficient to reach a final conclusion concerning the age of metamorphism of the RMC. The *Papuk metamorphic complex* was subjected to metamorphism and migmatitization during the Caledonian orogeny (Jamičić 1988). It consists predominantly of (a) S-type granites which are enveloped in the NE and SW by (b) migmatites and migmatitic gneisses which grade into (c) amphibolite facies metamorphic sequences composed of garnetiferous amphibolites, paragneisses and micaschists (Pamić 1986; Pamić & Lanphere 1991).

According to Jamičić (1988) the PTSS uncomformably overly the Paleozoic rocks, and is characterized by several lithostratigraphic units. The base of the sequence is built up by coarse clastic rocks represented by phyllitic conglomerates and sandstones which grades continuously into red to purple fine-grained sandstones and siltstones. This facies contains granitoid, gneiss and pegmatite clasts derived from the metamorphic complex of the Papuk Mountain. The second group, represented by fine-grained quartz sandstones, is transitional towards the Lower Triassic strata. The Lower Triassic sequence, as part of the PTSS, is represented by sandstones, calcareous sandstones and siltstones. Sedimentation in the area continued into the Middle Triassic which is represented by dolomites, calcareous-dolomitic breccias and subordinate limestones. Recently, Biševac et al. (2009) showed that the PTSS underwent very low- to low-grade metamorphism. This metamorphism affected predominately the clay minerals, thus leaving hardly noticeable marks and reassigning (in the strict sense of metamorphic classification) the Mesozoic sedimentary rock complex into very-low grade metasediments. Since the effects of metamorphism studied here are hardly or not at all visible without the aid of instrumental techniques, we will avoid declaring them as metasediments.

Analytical methods

Isotope geochronological measurements were preceded by petrographic investigations in order to select the appropriate samples representing the PMC, RMC and PTSS units for further analyses (Fig. 1). A detailed description of the petrographic features of the representative samples, as well as the procedure of sample preparation and the petrographic technique used were described in Biševac et al. (2009). To assure the possibility of an inter-laboratory correlation of the Kübler index (KI) and Árkai index (ÁI) some details of the X-ray diffraction (XRD) work are given here. For detailed explanation of the standardization procedure together with measurement conditions see Biševac et al. (2009).

The whole rock powder XRD analysis of the samples (a modal composition determination) was performed on a Philips X'Pert Pro diffractometer equipped with the X'celerator detector using CuKα radiation from a tube operating at 40 kV and 45 mA. The step width was 0.017° 2Θ with 43 s counting time per step; the samples were run between 4 and 65° 2Θ.

Special attention was given to the clay minerals (< 2 μm fraction) using the procedure proposed by Starkey et al. (1984). Samples were measured as highly oriented air dried preparations on glass slides additionally treated with ethylene-glycol (EG) and heated first at 400 °C and than at 550 °C. The instrumental settings were the same as for the modal composition determination.

KI and ÁI were measured after Kübler (1967, 1975, 1990) and Árkai (1991) respectively. The agreed-on boundary between the diagenetic zone and the anchizone at present is at KI = 0.42°Δ2Θ CuKα, while for the anchizone-epizone boundary it is KI = 0.25°Δ2Θ CuKα. These boundaries are associated with temperatures of approximately 200 °C and 300 °C respectively (Kübler 1968; Warr & Rice 1994). The anchizone is further divided into a high and low temperature anchizone. The boundary between these two zones is KI = 0.30°Δ2Θ CuKα and corresponds to the temperature of approximately 260 °C (Potel et al. 2006). The boundaries of the anchizone for ÁI were redrawn from Árkai et al. (1995b) being: ÁI (001) = 0.26–0.37°Δ2Θ and ÁI (002) = 0.24–0.30°Δ2Θ CuKα. Sample preparation was made according to the recommendation of Kisch (1991) and the full width at half maximum (FWHM) was read manually. KI and ÁI were measured on air dried and EG treated samples. No shift of the basal white mica reflection after EG treatment was observed, hence discussion is based only on the air-dried scan results. The standardization of the KI and ÁI values of samples measured in the laboratory to those from Kübler's laboratory, taken as reference values, was made using eight Kisch's standards, namely rock slabs polished parallel to the foliation (Kisch 1990; 1991). Crystallinity Index Standards (CIS) (Warr & Rice 1994) were used later for monitoring changes of the measured FWHM caused by tube ageing.

White K-mica geobarometry worked out for lower greenschist facies pelitic rocks by Sassi (1972) and extended by Padan et al. (1982) to the high temperature part of the anchizone was applied for qualitative estimation of the metamorphic pressure conditions. For these geobarometric estimations the constraints given by Guidotti & Sassi (1986) for appropriate modal composition were also taken into consideration. The b_0-parameter was measured on rock slabs cut perpendicular to the schistosity and, in order to avoid the influence of the detrital mica, on randomly oriented grain-size < 2 μm fraction using the same instrumental conditions as for KI and ÁI determination. The 2Θ range scanned was 59.0–63.0°, while quartz present within all analysed samples was used as the internal standard.

With the aim of correlating Kübler and Árkai indices with vitrinite reflectance, total organic carbon (TOC) was measured in order to find out whether the investigated samples were suitable for this kind of analyses. Samples were chosen on the basis of their colour, assuming that the darker samples should contain more organic matter. Sample preparation was done according to Bush (1970). Standardization of the instrument (LECO IR-212) was done using the material of a known carbon content (steel ring containing 0.3–1.0 % of carbon). For optical investigation of vitrinite particles a Leitz-MPV3 microscope was used. The standardization of the instrument was done using materials of known reflection index (sapphire, diamond and glass).

K-Ar dating was performed in the ATOMKI Institute of Nuclear Research, Hungarian Academy of Sciences. Illite–K-white mica fraction samples were degassed by high frequency induction heating; the released argon was cleaned by applying furnaces with Ti sponge and St707 getter materials. ^{38}Ar was introduced from a gas pipette. For Ar isotopic ratio measurements a magnetic mass spectrometer of 150 mm radius and 90° deflection was used in the static mode. Before the determination of K, the samples were digested by a mixture of $HF + HClO_4$ and dissolved in highly diluted HCl. The K content of the samples was measured with a flame emission photometer using 100 ppm Na buffer and Li internal standard. The results of an interlaboratory calibration were published by Odin et al. (1982). During this study interlaboratory standards of LP-6, HD-B1 and Asia 1/65 as well as atmospheric argon have been used for calibration. K-Ar ages were calculated using the constants proposed by the IUGS Subcommission on Geochronology (Steiger & Jäger 1977). K-Ar ages measured on < 2 μm (and in certain case whole rock) fractions were used for dating the metamorphism to avoid and/or reduce the disturbing effects of detrital muscovite following the practice of Clauer & Kröner (1979), Frank & Stettler (1979), Bonhomme et al. (1980), Reuter (1987) and Árkai et al. (1995a).

Results

The label, lithology, stratigraphic age and tectonic settings of the studied samples together with their semiquantitative modal composition, KI and ÁI values, b_0-parameter data, total organic carbon content, vitrinite reflectance value as well as K-Ar ages obtained on whole rock (WR), > 0.1 mm, < 2 μm and < 0.5 μm grain-size fractions are listed in Table 1.

Modal composition

As can be expected from the lithology, a semiquantitative whole rock modal composition of the samples as determined by the XRD method revealed similarities between the samples from the different tectonic units (Table 1). The dominant constituents of samples belonging to the RMC are quartz, illite-muscovite, plagioclase and chlorite. Less abundant minerals are K-feldspar and paragonite (Table 1). Plagioclase, augite and actinolite were the main constituents of the metadolerite from the RMC. The reddish colour of samples from all complexes (Table 1) is due to the presence of hematite in the matrix. Calcite, as a secondary mineral, was detected in slate from the RMC. Chloritoid and pyrophyllite, in association with illite-muscovite, quartz and subordinate chlorite is a characteristic of Upper Devonian samples found along the Kutjevo transect (Table 1). Kaolinite was detected as a trace mineral in metasandstones belonging to the PTSS in which main constituents are quartz and illite-muscovite.

Additional attention was focused on the clay minerals association. Although a mineral size should not be the only criterion defining a certain mineral group, the term "clay minerals" in this work refers to the sample < 2 μm fraction. The most abundant minerals in this fraction, detected by XRD on highly oriented mounts, were illite-muscovite, chlorite and quartz. Paragonite, pyrophyllite, plagioclase, hematite and kaolinite were occasionally observed and related to samples in which they were also detected in whole rock samples. Special interest was given to illite-muscovite and chlorite in order to determine possible interstratifications and because these minerals were used later for KI and ÁI analyses.

Kübler and Árkai indices

There is no significant difference between KI and ÁI values measured on samples from the different lithostratigraphic units (Table 1). The values point to high anchizonal to epizonal thermal alteration.

Total organic carbon (TOC) and vitrinite reflectance

The TOC in all analysed samples was absent or very low (Table 1). Nevertheless, vitrinite particles suitable for vitrinite reflectance measurements were found in one slate from the RMC (Table 1). The result (R_{random} = 4.59; n = 14, standard deviation = 0.217) indicates meta-anthracite coal rank.

b_0-parameter

The b_0-values evaluated from whole rock range between 9.000 and 9.036 Å, giving an average value of 9.021 Å, while those obtained on < 2 μm fraction vary between 8.986 and 9.036 Å, giving an average of 9.004 Å (Table 1).

K-Ar age dating

The K-Ar ages of different fractions are shown in Table 1. The oldest age was obtained from the WR of sample no. 4 (153.1 ± 6.6). The youngest age was obtained on the < 0.5 μm fraction of sample no. 34 (66.2 ± 2.5 Ma). The results together with analytical and statistical parameters and errors of the K-Ar isotopic measurements can be obtained from the corresponding author upon request.

Discussion

Clay mineralogy and P-T evolution

The generally accepted theory is that illite is formed by transformation of smectite as a result of heating (Hower et al. 1976; Merrimann & Peacor 1999). In spite of the fact that different authors propose different mechanisms of transformation of smectite to illite (Morton 1985; Dong 2005), all authors agree that the effect of heating in the presence of a K-containing fluid will result in progressive loss of smectite layers and a simultaneous increase of illite layers in interstratified illite-smectite.

Many researchers tried to assign a particular temperature to the thermal stability of smectite — interstratified illite-smectite — illite series. According to Weaver (1989), 5–10 % of smectite layers are present in illite-smectite interstratified clays at 200–250 °C, while according to Viczián (1994) around 50 % of smectite layers are present at 90–130 °C and 5–10 % at 160–220 °C. Inoue et al. (2004) gave the most definite statement: smectite is stable from room temperature up to 150 °C, the R1 type of illite-smectite interstratification in the range of 150–225 °C, while illite is stable at temperatures higher than 175 °C.

In order to determine a possible presence of smectite, either as a discrete mineral phase or interstratified with illite, the classical XRD analysis as proposed by Starkey et al. (1984), was performed. After these treatments, no shift of the 10 Å diffraction maximum of illite-muscovite and the 14 Å diffraction maximum of chlorite was observed (Fig. 2). The data, characteristic for all analysed samples, imply that the samples do not contain smectite. In order to ascertain this statement the concept of poorly- (PCI) and well-crystallized illite (WCI)

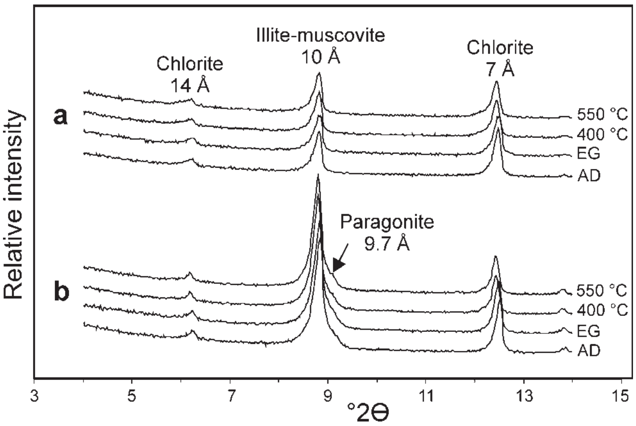

Fig. 2. Representative XRD patterns pertaining to: **a** — phyllite no. 13 and **b** — metasandstone no. 4. Both samples belong to the RMC. **AD** — air dried; **EG** — ethylene-glycol treated mounts.

was used. In general, as the number of coherent diffracting domains becomes smaller, the diffraction peak becomes wider and shifts to higher *d*-values. The width of peak at half height (FWHM) and the position of the peak maximum of the studied samples were plotted in the appropriate diagram (Lanson et al. 1998) (Fig. 3). The position of the studied samples in the diagram and lack of shift to higher *d*-values after EG treatment indicates an absence of interlayering even with very small amounts of smectite. The data point to thermal alteration higher than 220–250 °C, taking into consideration the smectite thermal stability pointed out by Weaver (1989), Viczián (1994) and Inoue et al. (2004). For better estimation of the degree of thermal alteration, KI and ÁI were used. Although these parameters are very good tools for monitoring the progress of the phyllosilicate reaction and their transformation with increasing temperature, the exact temperature of thermal alteration can only be estimated. KI indicates that the samples

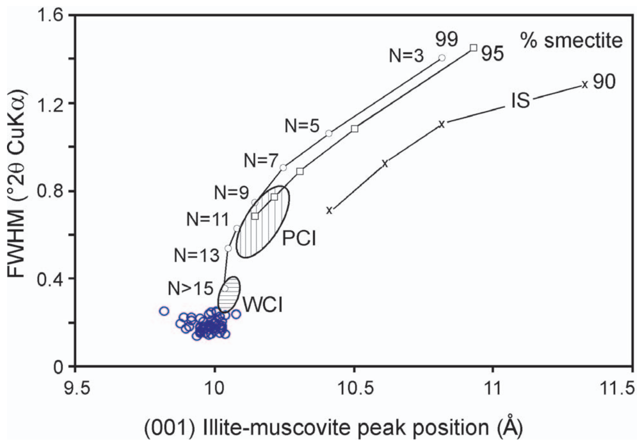

Fig. 3. Peak positions and full width at half maximum (FWHM) of the (001) diffraction maximum of illite-muscovite shown in the diagram of Lanson et al. (1998). *N* indicates number of diffracting layers. The shaded areas indicate poorly- (PCI) and well-crystallized illite (WCI) obtained from sequences of sedimentary rocks reported by Lanson et al. (1998).

Table 1: An overview of rock type, stratigraphy, semiquantitave mineral composition (●● — dominant; ● — abundant; ○ — significant; x — poor), "crystallinity" values, b_0-parameter, total organic carbon (TOC) content, vitrinite reflectance (R_{random}) and K-Ar ages of samples representing the Psunj metamorphic complex, the Radlovac metamorphic complex and the Permian-Triassic sedimentary sequences. * — ÁI (002) determined after treatment with DMSO; ** — according to Jamičić & Brkić (1987); *** — calculated with atomic constants suggested by Steiger & Jäger (1977). Legend: **PCm** — Precambrian; $\mathbf{D_3}$ — Upper Devonian; **C** — Carboniferous; **P** — Permian; **PT** — Permo-Triassic; **T** — Triassic. The mineral abbreviations are after Kretz (1983): **Qtz** — quartz; **Ill-Ms** — illite-muscovite; **Pl** — plagioclase; **Kfs** — K-feldspar; **Chl** — chlorite; **Cld** — chloritoid; **Prl** — pyrophyllite; **Hem** — hematite; **Kln** — kaolinite; **Pg** — paragonite; **Act** — actinolite; **Aug** — augite; **Cal** — calcite.

Sample	Rock type	Stratigraphy**	Modal composition													KI (001)	ÁI (001)	ÁI (002)	b_0 parameter (Å)		TOC (%)	R_{random} (%)	K-Ar ages (Ma)***			
			Qtz	Ill-Ms	Pl	Kfs	Chl	Cld	Prl	Hem	Kln	Pg	Act	Aug	Cal		°Δ2Θ		whole rock	<2 µm			>0.1 mm	whole rock	<2 µm	0.5 µm
Psunj metamorphic complex (PMC)																										
34	chlorite schist	PCm	●	●	○		●●									0.244	0.243	0.265						143.4 ± 5.4	83.8 ± 3.2	66.2 ± 2.5
72	schist	PCm	●	●●	●		○									0.259		0.258								
Radlovac metamorphic complex (RMC)																										
36	metagreywacke	D_3	○	○	x		○	●	x	x						0.321	0.296	0.274								
37	metagreywacke	D_3	○	●	x		x	●	x	x						0.305		0.306								
38	metagreywacke	D_3	●	○	x		○	●●								0.327		0.291								
39	metagreywacke	D_3	●●	○	x		x	●	x							0.271										
40	chloritoid schist	D_3	●	●	x	x	x	●								0.290					0.13			112.6 ± 4.4	89.7 ± 3.5	74.5 ± 2.8
43	chloritoid schist	D_3	○	●	x	x	x	●	○							0.292								113.6 ± 4.3	83.1 ± 3.3	81.5 ± 3.3
59	graphitic schist	D_3	●●	●	○		●									0.247	0.254	0.246								
57A	metasandstone	C, P	●●	●	○	x	○										0.273	0.269								
57C	metasandstone	C, P	●●	○	●		○					x					0.309	0.277								
57D	metasandstone	C, P	●●	○	●		○					x				0.306	0.266	0.267								
60	metasandstone	C, P	●●	●	○		○										0.292	0.281								
61	metasandstone	C, P	●	●●	○		○			x							0.288	0.273								
62	metasandstone	C, P	●	●●	○		○			x						0.269	0.273	0.270								
1A	metasandstone	C, P	●	●	○		●					x				0.247	0.241	0.224	9.017	8.995	0.02					
1B	slate	C, P	●	●	○		●					x				0.249	0.234	0.241	9.029	9.008	0.02		180.0 ± 6.8	138.6 ± 5.3	94.0 ± 3.8	79.9 ± 3.1
2	phyllite	C, P	●	●	○	x	●									0.240	0.253	0.240			0.00					
3	phyllite	C, P	○	●	○	x	●●									0.239	0.236	0.234	9.010	8.980	0.10					
4	quartzite	C, P	●●	●	○		○					x				0.286	0.223	0.224					256.7 ± 10.5	153.1 ± 6.6	92.3 ± 3.6	79.9 ± 3.1
5	slate	C, P	●	●●	○	x	○									0.239	0.231	0.234								
6A	phyllite	C, P	●	●●	○		○			x						0.219	0.210	0.215								
6B	phyllite	C, P	●	●●	○		●									0.244	0.225	0.236			0.00					
7	slate	C, P	●	●●	○	x	○									0.250					0.02					
8	metaconglomerate	C, P	●	●●	x	x	○			x						0.238	0.221	0.232			0.00					
9	phyllite	C, P	●	●●	x		○			x		x				0.253		0.242	9.000	8.996	0.01			122.1 ± 4.6	99.3 ± 3.9	88.2 ± 3.4
10	phyllite	C, P	●	●●	x		○			x		x				0.265	0.233	0.230								
11	metasandstone	C, P	●	●●	○		●									0.243	0.291	0.259			0.20					
12	slate	C, P	●●	●	○		○								x	0.249	0.259	0.238	9.023	8.986	0.12	4.59				
13	phyllite	C, P	●	●●	x		●									0.315	0.224	0.228			0.60					
14	slate	C, P	○	●●	○	x	○			x						0.223	0.220	0.224			0.02					
15	phyllite	C, P	●	●●	○	x	○			x						0.265	0.281	0.263	9.026	9.030						
16	slate	C, P	●	●●	○		○			x						0.258	0.261	0.252								
17	phyllite	C, P	●	●●	○		○			x						0.253	0.250	0.246	9.032	9.036						
18	slate	C, P	●●	●	○		○			x						0.262	0.277	0.257	9.036	9.035						
19	phyllite	C, P	●	●●	x		x			○		x				0.332		0.302								
23	siltstone	C, P	●●	●			x	○				○						0.276								
24	siltstone	C, P	●	●●	○		○	x								0.278		0.278	9.011	8.992						
25	phyllite	C, P	●	●●	○		○			x						0.255	0.260	0.255								
31	phyllite	C, P	●	●●	○		●	●											9.029	9.000						
32	metasandstone	C, P	●●	●	○		○									0.342										
33	phyllite	C, P	●	●	○		●●	●											9.023	8.990						
63	metaconglomerate	C, P	●	●●	x	x	○			x						0.347		0.310								
64A	slate	C, P	●	●●	x		○			x		x				0.265	0.265	0.235								
64B	slate	C, P	●	●●	x		●					x				0.242	0.273	0.234								
65	slate	C, P	●●	●	○		●									0.205	0.245	0.203								
66	slate	C, P	●	●●	○		●									0.225										
67A	metadiabase	C, P	x	x	●●		●						○	x			0.238	0.218								
67B	slate	C, P	●●	●	○		○			x						0.213	0.254	0.222								
67C	slate	C, P	●●	●	○		○			x						0.207	0.308	0.221								
67D	metaconglomerate	C, P	●	●●	x	x	○			x						0.242										
68A	phyllite	C, P	●●	●	○		●									0.294	0.336	0.285								

Table 1: *Continued*

Sample	Rock type	Stratigraphy**	Modal composition													KI (001)	ÁI (001)	ÁI (002)	b_0 parameter (Å)		TOC (%)	R_{random} (%)	K-Ar ages (Ma)***			
			Qtz	Ill-Ms	Pl	Kfs	Chl	Cld	Prl	Hem	Kln	Pg	Act	Aug	Cal		°Δ2Θ		whole rock	<2 µm			>0.1 mm	whole rock	<2 µm	0.5 µm
68B	phyllite	C, P	●	●●	○		●									0.221	0.224	0.205								
68C	slate	C, P	●●	●	○		○									0.245	0.239	0.218								
68D	metaconglomerate	C, P	●	●●	○		●									0.253		0.249								
69	slate	C, P	●	●	○		○									0.235	0.300	0.221								
70A	metaconglomerate	C, P	●	●●	○		●									0.302	0.312	0.242								
Radlovac metamorphic complex (RMC)																										
70B	slate	C, P	●	●●	○		●									0.244	0.262	0.236								
71A	phyllite	C, P	●●	●	○		●									0.237	0.267	0.226								
71B	slate	C, P	●	●●	○		○									0.265	0.348	0.281								
Permian-Triassic sedimentary sequences (PTSS)																										
44	phyllitic metaconglomerate	[1]PT	●	●●	○		○			x						0.238		0.283						120.8 ± 4.6	85.0 ± 3.2	83.1 ± 3.1
46	phyllitic metaconglomerate	[1]PT	●	●●	○		○			x											0.00					
47	phyllitic metaconglomerate	[1]PT	●	●●	○		○			x						0.262	0.221	0.232						109.0 ± 4.1	79.4 ± 3.1	72.8 ± 2.8
58	phyllitic metaconglomerate	[1]PT	●●	●	x		○			x		x				0.299		0.273								
49	quartz metasandstone	[2]PT	●	●●		○	○				x					0.248		0.274*			0.00			130.0 ± 4.9	91.6 ± 3.3	78.0 ± 3.3
50	metasandstone	T_1	●●	●	○	○					x										0.00			86.0 ± 3.3	71.4 ± 2.8	71.7 ± 3.3
53	metasandstone	T_1	●●	●	x	x					x					0.259										
56	metasandstone	T_1	●●	●	x	●										0.337										

passed through transitional high anchizonal to epizonal thermal alteration (Table 1, Fig. 4). Similar results were obtained using ÁI (Table 1, Fig. 4). KI and ÁI (002), as well as ÁI (001) and ÁI (002) show a good correlation (Fig. 5). The estimated degree of thermal alteration is in good agreement with the previously established presence of paragonite and pyrophyllite in some slates from the RMC as reported by Slovenec (1986). According to Frey (1987) these two minerals are characteristic of the sub-greenschist facies and indicate, as well as KI and ÁI measured here, that the RMC experienced a very low- to low-grade metamorphic event. The data indicate that samples from the PMC as well as the overlying PTSS sampled along the Kutjevo transect also record a very low- to low-grade metamorphic event. The degree of metamorphism appears to be constant in all the analysed samples irrespective to their stratigraphic age.

It is important to mention that small differences between KI and ÁI data are closely related to the chlorite content. Lower chlorite content results in weaker basal reflections and consequently less precise measurement of the FWHM.

The presence of kaolinite in the samples can make ÁI (002) measurements very difficult and uncertain because of the overlapping of the (001) diffraction maximum of kaolinite and the (002) diffraction maximum of chlorite. This problem can be overcome by the dimethyl-sulfoxide (DMSO) treatment as shown in the study of Biševac et al. (2009). The appearance of ordered kaolinite in some samples (Table 1) can be connected to post-metamorphic hydrothermal alteration as proposed by Árkai et al. (2000).

Good correlation of KI and ÁI for samples containing paragonite or pyrophyllite indicates that they did not have any significant influence on the measured KI values due to their low quantity.

In order to correlate the KI and ÁI with other parameters which change with the thermal conditions, vitrinite particles were measured on one slate sample from the RMC. The vitrinite reflectance corresponds to meta-anthracite coal rank. The value is in good agreement with the KI and ÁI for the same sample (Table 1).

Epizonal (chlorite zone) and medium- to high-temperature anchizonal fine-clastic metasedimentary rocks with common mineral assemblages consisting of quartz, albite, white K-mica±chlorite and calcite can be taken into consideration for evaluating pressure conditions using the b_0-parameter (Padan et al. 1982; Guidotti & Sassi 1986). The data for <2 µm grain-size fraction implies transitional low- to medium-pressure formation conditions for RMC, while whole rock measurements on same samples indicate medium pressure conditions. The b_0-parameter of the <2 µm fraction can be related to the metamorphic pressure while the slightly higher pressure conditions indicated by whole rock measurements can be explained by the influence of the detrital mica.

We conclude that the analysed samples, regardless to their stratigraphic age, record the same metamorphic event which is indisputably younger than Early Triassic, that is, younger than their protolith ages. Middle Triassic dolomites and dolomitic limestones (Jamičić & Brkić 1987; Jamičić et al. 1987), representing the youngest rocks of the Kutjevo transect, were omitted from this research because of the lack and/or very low

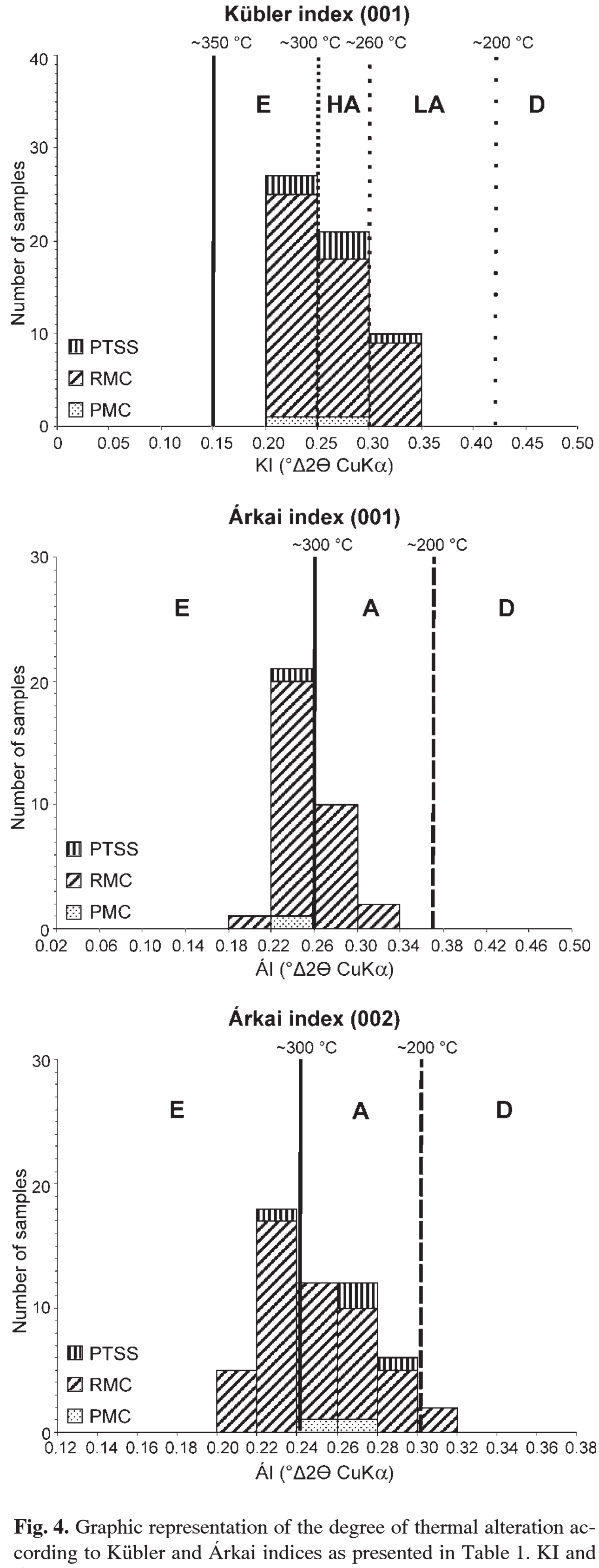

Fig. 4. Graphic representation of the degree of thermal alteration according to Kübler and Árkai indices as presented in Table 1. KI and ÁI values are expressed in °Δ2Θ CuKα. Boundaries of the anchizone were taken from Kübler (1968) and Potel et al. (2006) for KI and from Árkai et al. (1995b) for ÁI. "E" — epizone; "HA" — high anchizone; "LA" — low anchizone; "A" — anchizone;"D" — diagenetic zone.

quantity of illite-muscovite which can influence the reliability of the KI measurements. Upper Jurassic-Lower Cretaceous sequences, otherwise present in the Slavonian Mts, cannot be found in the investigated area.

Chronology of metamorphism

When evaluating and interpreting the K-Ar data in order to determine the age of the metamorphism, a possible influence of the detrital material should be taken into consideration (Hunziker et al. 1986). The effect of the detritus was tested by dating different mineral grain fractions (<0.5 μm and <2 μm) including the whole rock. The whole rock age is the oldest one in all cases (Table 1). The youngest age was measured on <0.5 μm fractions. The whole rock age represents the average age of all K-bearing constituents present in the rock, while the age of finer fractions, <0.5 μm and <2 μm in this case, corresponds to the average age of the different generations of illitic material present within the dated fractions. Such interpretation suggests that the oldest K-Ar age constrains the lowest age for the metamorphic event which reset most of the mineral ages in the dated rocks. The youngest age, measured on the <0.5 μm fraction, sets the oldest limit for the termination of illite formation. The K-Ar whole rock age does not have any significant geological meaning, it only implies that minerals older and younger than the whole rock age exist. A microscopic investigation revealed that those minerals could be coarse-grained mica flakes (Fig. 6.1–3). The K-Ar ages of coarse-grained muscovite (>0.1 mm fraction) confirmed this assumption (Table 1). Concerning the well known closure temperature concept of the K-Ar system (Purdy & Jäger 1976; Wagner et al. 1977), the results indicate that the formation of illite minerals took place at temperatures below the blocking temperature of muscovite (~350 °C; Rollinson 1993). If this was not the case, the coarse-grained muscovite would be fully reset. Such an interpretation is in good agreement with the estimates of the degree of thermal alteration deduced from the KI and ÁI data.

As recommended by Árkai et al. (1995a, 2000), the <2 μm fraction ages were used for determination of the metamorphic age. The exception was Early Triassic metasandstone (PTSS) which, according to the microscopic investigation, does not contain coarse-grained detrital mica (Fig. 6.4). We can assume that the thermal alteration that affected this lithostratigraphic unit reset all the K-bearing minerals present in it. The observed difference between K-Ar whole rock age and ages of the finer fractions (in this case <0.5 μm and <2 μm) is the consequence of prolonged formation of clay minerals. For this reason, the K-Ar whole rock age of Early Triassic metasandstone, assumed to be equivalent to the <2 μm ages of other samples which contain coarse-grained mica, was used for discussion regarding the age of the metamorphism.

If we take into consideration that in all dated samples, ranging from Early Paleozoic to Early Triassic, the K-Ar age data of clay fractions point to Late Cretaceous, then the age of the very low- to low-grade metamorphic event can be set as Eoalpine (Table 1, Fig. 7). On a regional scale this event is most likely related to the main nappe-forming compressional events in the Pannonian Basin area and the Carpathians which result-

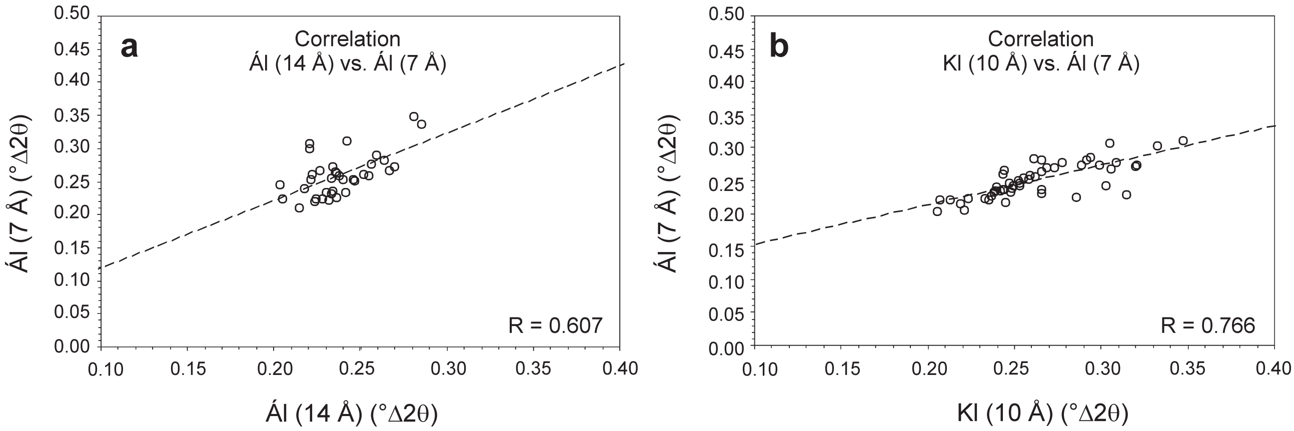

Fig. 5. Correlation of Kübler index and Árkai index as metamorphic indicators: **a** — ÁI (14 Å) vs. ÁI (7 Å); **b** — KI (10 Å) vs. ÁI (7 Å).

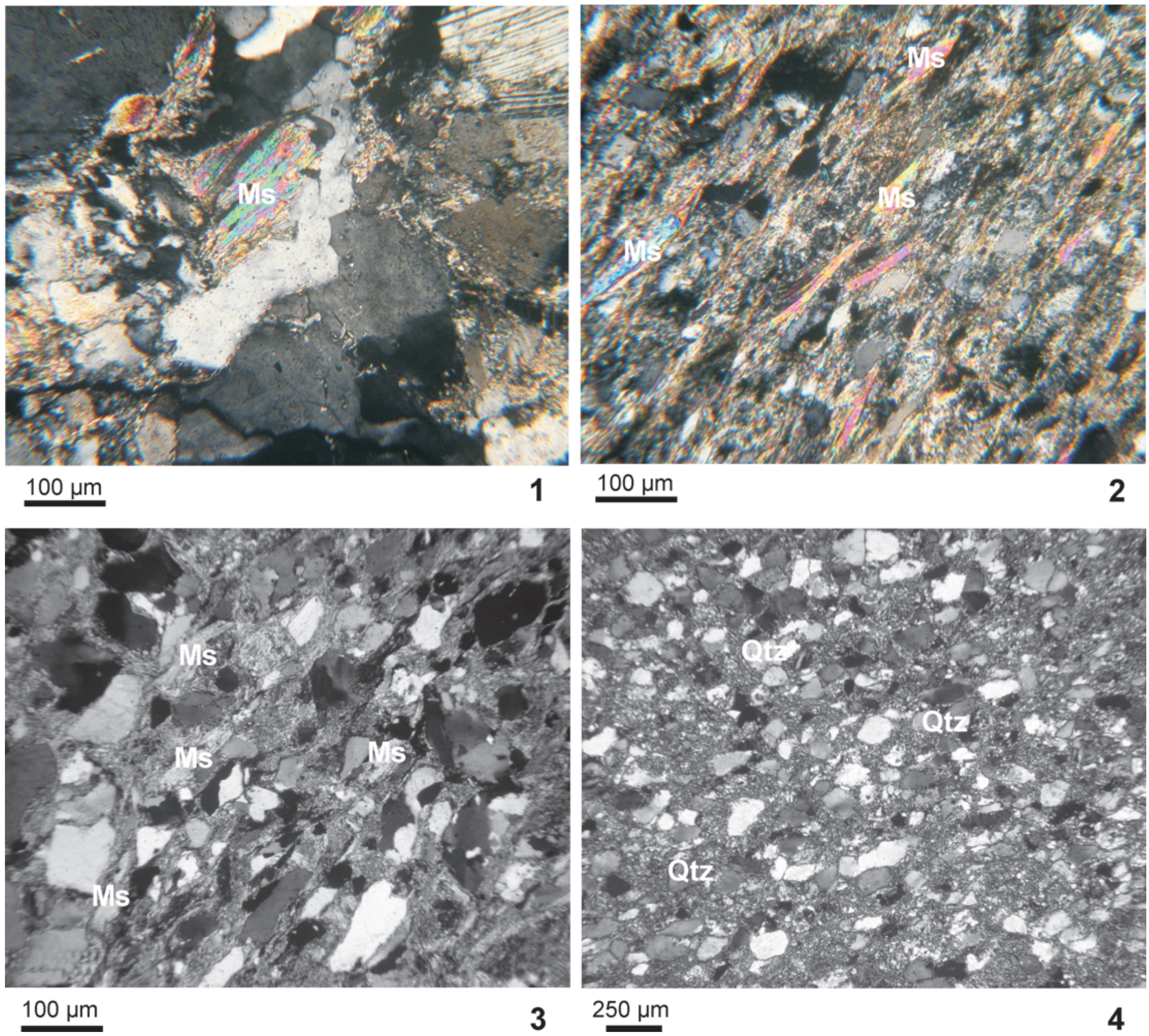

Fig. 6. Microphotographs showing samples of: **1** — phyllite (RMC, no. 1B); **2** — quartzite (RMC, no. 4); **3** — metasandstone (RMC, no. 32) in which coarse-grained muscovite flakes are clearly visible; **4** — Metasandstone (PTSS, no. 50) does not contain coarse-grained mica. The mineral abbreviations are after Kretz (1983): **Qtz** — quartz; **Ms** —muscovite.

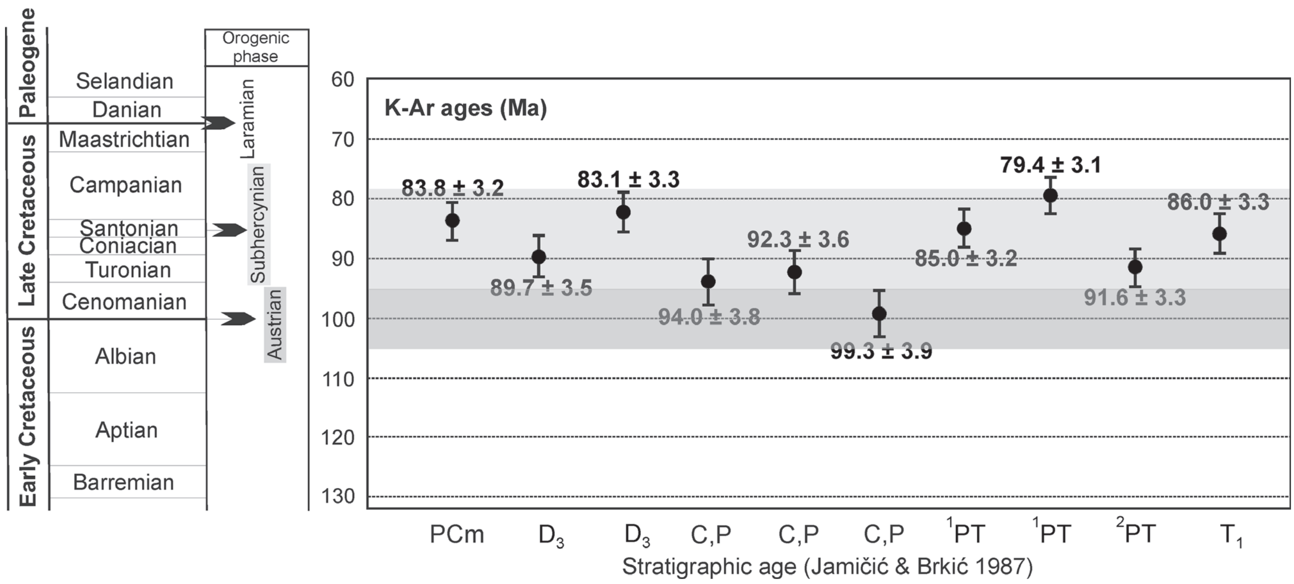

Fig. 7. Graphic representation of the K-Ar ages measured on <2 μm fraction. Whole rock age of sample no. 50 (T_1; metasandstone; PTSS) is shown (see text for detailed explanation). Shaded areas represent the approximate duration of Alpine orogenic phases.

ed in the nappe stacking. Similar results have been already reported from other parts of Tisia and ALCAPA (Árkai 2001 and references therein). The Late Cretaceous deformational event, distinctive for the Tisia and Dacia area, can be distinguished from the Early Cretaceous deformational episode recorded in the ALCAPA and Dacia (Schmid et al. 2008).

Earlier research (Jamičić 1988) associated the metamorphism of the RMC with the folding processes during the last stages of the Variscan orogeny. According to present knowledge there is no reliable age dating proving Variscan metamorphism of the RMC. Moreover most of the RMC samples of Carboniferous to Permian age have one clearly visible schistosity. Nevertheless the presence of Variscan metamorphism of the RMC with a grade not higher than that of the recorded Cretaceous event cannot be ruled out unequivocally. Reasons for this opinion are the presence of two foliations on some samples from the RMC and the presence of differently oriented clasts of very low- to low-grade metamorphic rocks in the Permian-Triassic metaconglomerates. Nevertheless, our K-Ar data (Table 1) indicate that rocks belonging to the PMC, RMC and PTSS were altered during the Cretaceous. No considerable systematic or gradual variation between the K-Ar age of the <2 μm fraction and metamorphic indicators of the analysed samples, regardless of the stratigraphy, was observed (Fig. 8). This could imply that the analysed tectonic units were not additionally affected by younger thermal alterations.

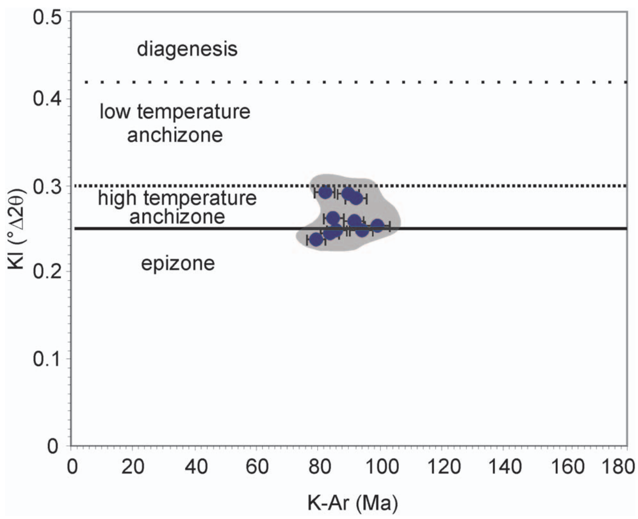

Fig. 8. Correlation of KI values with the K-Ar ages of <2 μm fraction. For sample no. 50 (T_1; metasandstone; PTSS) whole rock age is shown.

Correlation with the surrounding areas of the Tisia Unit and ALCAPA

Most of the previous researches regarding the metamorphic evolution, using Kübler and Árkai indices, b_0-parameter and K-Ar dating, are related to the Paleozoic and/or Mesozoic sequences of the Hungarian part of the Tisia Unit, but also to the surrounding area belonging to ALCAPA (Árkai 1977, 1983, 1995; Árkai et al. 1981, 1995a, 1998, 2000, 2003; Sadek-Ghabrial et al. 1996; Faryad & Henjes-Kunst 1997; Janák et al. 2001; Lupták et al. 2003). Similar very low-grade metamorphic studies of the Croatian part of Tisia are rare. We compare our results with those from similar studies in other parts of the Tisia Unit (Mecsek, Villány-Bihor and Békés-Codru nappe systems), the Bükkium (Bükk, Uppony and Szendrő) and parts of the Central Western Carpathians (Veporic and Gemeric Units) (Table 2).

A close connection between the metamorphic evolution of the Hungarian and Croatian parts of the Tisia can be established according to the data presented in Table 2. All Paleozoic or Mesozoic rocks of the Tisia Unit were affected by an

Table 2: Data from this work (Kübler and Árkai indices, vitrinite reflectance, b_0-parameter and K-Ar dating) as compared with data from other parts of Tisia (Mecsek, Villány-Bihor and Békés-Codru) and ALCAPA (Bükkium, Veporic and Gemeric).

Mega-Unit	TISIA (Croatia)	TISIA (Hungary)			ALCAPA (Hungary)			ALCAPA (Slovakia)	
Tectonic Unit		Nappe system			Western Carpathians			Central Western Carpathians	
					BÜKKIUM				
Metamorphic indicator	Radlovac metamorphic complex	Mecsek	Villány-Bihor	Békés-Codru	Bükk	Uppony	Szendrő	SUPERUNIT VEPORIC	SUPERUNIT GEMERIC
Thermal alteration (Kübler and Árkai indices)	High anchizonal to epizonal	Anchizonal to epizonal	Late diagenesis to anchizonal	Anchizonal, transitional anhi- to epizonal, partly epizonal	Anchizonal	Transitional anhi- to epizonal	Epizonal	Transitional anhi- to epizonal	High anchizonal
Vitrinite reflectance	High anchizonal to epizonal	Anchizonal to epizonal	Data not available	Data not available	Data not available	Transitional anhi- to epizonal	Greenschists facies	Data not available	Data not available
Pressure conditions (b_0-parameter)	Transitional low to intermediate	Low to intermediate	Transitional low-intermediate to intermediate	Lower intermediate	Low to intermediate	Low	Low to intermediate	Intermediate (4–4.5 kbars)	Intermediate (2.5–5 kbars)
Age of metamorphism (K-Ar dating)	Late Cretaceous		Late Cretaceous		Cretaceous	Cretaceous	Cretaceous	Early Cretaceous	Early Cretaceous Late Cretaceous
Comment(s)	• Late Cretaceous high anchizonal overprint of PMC • Late Cretaceous high anchizonal metamorphism of PTSS		• High anchizonal to epizonal overprint of older polymetamorphic complex parallel to prograde metamorphism of Mesozoic formation			• Only one, Alpine, metamorphic event was established		• Chlorite thermometer indicates ~310–380 °C • Pressure was estimated using THERMOCALC software	• Evidence of the Late Cretaceous (~90 Ma) overprint was established by Ar-Ar spectra
References	Pamić & Lanphere (1991); Jerinić et al. (1994); Biševac et al. (2009).		Árkai et al. (1998, 2000); Árkai (2001 and references therein)			Árkai (1977, 1983), Árkai et al. (1981, 1995a), Sadek-Ghabrial et al. (1996)		Jának et al. (2001); Lupták et al. (2003)	Faryad & Henjes-Kunst (1997); Árkai et al. (2003)

Eoalpine (Cretaceous) metamorphic event. Where measurements were possible, vitrinite reflectance data correlate well with the values of "crystallinity" indices. A similar situation regarding the P-T evolution can be observed by comparing the Tisia and surrounding area belonging to the ALCAPA (Table 2). While the Tisia Unit is characterized only by the Late Cretaceous very low- to low-grade metamorphism, in the ALCAPA region, as well as in Dacia, the additional Early Cretaceous metamorphic event, which did not affect Tisia, can be distinguished (Schmid et al. 2008).

Conclusions

1. The XRD analysis of clay minerals indicates the presence of well-crystallized illite (WCI) in all samples. The ordered crystal structure of illite points to a thermal alteration of at least 220–250 °C. Chlorite, present in the clay fraction, also represents a stable mineral phase indicating anchizonal to epizonal thermal alteration.

2. The Kübler index and Árkai index show good correlation and indicate that samples passed through a high-anchizonal to epizonal thermal alteration. Variation of KI and ÁI data with the stratigraphic ages was not observed. All samples recorded the same metamorphic conditions.

3. The total organic carbon (TOC) content in all samples is very low. Nevertheless, vitrinite reflectance data, measured in a single sample, indicates a meta-anthracite coal rank and are in accordance with both mineral composition and KI and ÁI data.

4. Pressure conditions estimated on the basis of the b_0-parameter indicate that a metamorphic alteration of samples from the RMC proceeded in a transitional low-medium pressure system.

5. K-Ar dating of different grain size fractions of investigated samples revealed decrease in age with decreasing grain size of the dated fraction. The whole rock age is the oldest, while the youngest age is always obtained on the <0.5 µm fraction. This effect is closely connected to the amount of detrital mica in the fraction. Additional K-Ar dating revealed that the oldest K-bearing phase in analysed samples is detrital muscovite, giving an age which is older than the whole rock age. This indicates that the formation of illite minerals took place at temperatures that are lower than the closure temperature of muscovite (~350 °C). The presence of detrital muscovite implies an easy access to K, which may help the prolonged low-temperature formation of fine-grained illite.

6. K-Ar ages of <2 µm fraction indicates Late Cretaceous metamorphism of the PMC, RMC and PTSS.

7. Correlation of K-Ar ages with other metamorphic indicators (KI and ÁI) relating to stratigraphic age was not observed.

8. K-Ar data imply that all the analysed samples were altered as a consequence of a very low- to low-grade metamorphic event presumably related to the main nappe-forming compressional events in the Pannonian Basin and the Carpathians.

9. The investigation of P-T-t evolution in a certain timeframe presented here is in good agreement with similar researches conducted in Hungary (Mecsek, Villány-Bihor and Békés Codru) showing the same Eoalpine metamorphic evolution of the Hungarian and Croatian parts of the Tisia Megaunit.

Acknowledgments: We thank Prof. H.J. Kisch from the Department of Geology and Mineralogy, Ben-Gurion University of Negev, Beer-Sheva, Israel and Prof. Warr from Geologisch-Paläontologisches Institut, Ruprecht Karls Universität, Heidelberg, Germany for providing a set of standards necessary for instrument calibration in this study. We also thank Goran Šutej for measuring "crystallinity" of same samples as part of his diploma thesis and Darko Španić from INA — Oil Company, Corporate Processes, Research and Development Sector for measuring TOC and vitrinite reflectance. The authors would also like to thank reviewers Prof. P. Árkai and Prof. W. Frisch for their stimulating comments. This study was supported by the Ministry of Science, Education and Sports, project no. 119-1191155-1156 and by the Hungarian Academy of Science, Project No. OTKA T060965.

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