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Radiometric K/Ar data as evidence of the geodynamic evolution of the Ždraljica Ophiolitic Complex (central Serbia)

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Abstract. The study presents age data and petrologic characteristics of igneous rocks of the Ždraljica ophiolitic complex (ŽOC), situated in central Serbia, 150 km south of Belgrade. The complex consists predominately of a MORB/VAB-like tholeiitic suite, represented mostly by gabbros and diabases. The tholeiitic suite is intruded by calc-alkaline intermediate and acid magmas of a VA-affinity, which presumably formed in a pre-collisional setting. The whole complex is intruded by peraluminous granite magmas. The crystallization age of the calc-alkaline pre-collisional quartzdiorite is 168.4±6.7 Ma and it post-dates the formation of the here exposed oceanic crust. Geological evidence suggest that the emplacement of the complex occurred during the Upper Jurassic. With respect to their petrology and age, the Ždraljica ophiolitic rocks are similar to the south Apuseni Mts. ophiolites, situated to the north, and to the Kuršumija and Guevgeli ophiolites, situated to the south. All these ophiolites probably formed as parts of a single Jurassic belt, which can be termed the eastern branch of the Vardar Zone.

Key words: Vardar zone, Jurassic, ophiolites, K/Ar data, geodynamic, subduction, collision.

Апстракт. У овом раду приказани су подаци о старости и о најважнијим петролошким одликама магматских стена офиолитског комплекса Ждраљице који се налази у централној Србији, 150 km јужно од Београда. Комплекс је изграђен претежно од толеитских стена MORB/VAB карактера које су представљене углавном габровима и дијабазима. Ову толеитску свиту интрудују калко-алкалне интермедијарне и киселе магме VA-афинитета, за које се претпоставља да су образоване у пре-колизионим условима. Цео комплекс је интрудован пералуминијским леукократним гранитима. Време кристализације калко-алкалног кварцдиорита је 168.4 ± 6.7 Ма и оно највероватније указује на пре-колизиони стадијум. Геолошки докази указују да је офиолитски комплекс Ждраљице смештен током горње јуре. Према петрологији и старости, испитивани комплекс показује велику сличност са офиолитима јужних Апусена на северу и са офиолитским комплексима Куршумлије и Ђевђелије на југу. Сви ови офиолити су вероватно формирани као делови једног јурског појаса који данас припада источном ободу Вардарске зоне.

Кључне речи: Вардарска зона, јурски офиолити, K/Ar подаци, геодинамика, субдукција, колизија.

Introduction

Ophiolitic complexes are remnants of the oceanic lithosphere which where obducted onto colliding continental plates. They give precious information about the composite evolution of the oceanic crust/lithosphere, including spreading, lateral movements and closure of oceanic realms by subduction and obduction processes. These phases of the oceanic evolution were followed by metamorphic processes which usually make the investigation of magmatic processes very difficult.

The various ophiolites of the central and western Balkan Peninsula, mostly occurring in Serbia and Bosnia, and southward to the F.Y.R. of Macedonia and Greece, belong to a planetary ophiolitic zone connecting the Alpian and Himalayan orogenic belts (e.g. Coleman, 1977). Their nature has been a matter of ongoing debate, especially regarding the number of inferred paleo-oceanic domains which existed during the Mesozoic in this region. Most authors generally distinguish Dinaride and Vardar Zone ophiolites. They are regarded as belonging to separate sutures, situated in the west

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and in the east of the Drina–Ivanjica–Pelagonian block, respectively (e.g. KARAMATA *et al.*, 1999, KARAMATA *et al.*, 2000b, RESIMIĆ-ŠARIĆ *et al.*, 2000). While there is consensus for the Jurassic age of the emplacement of the Dinaride ophiolites, the ophiolites of the Vardar Zone are still the subject of various interpretations. For a long time the Vardar Zone ophiolites were regarded as being entirely Jurassic in age (ĆIRIĆ, 1996). During the last decades, evidence appeared that the Vardar Zone itself is a composite suture. This conclusion was mainly derived from geological observations that the ophiolites of the western part of the Vardar Zone are connected to a Sennonian olistostrome mélange (KARAMATA *et al.*, 2005 and references therein), but those in the eastern part are covered by transgresive Tithonian limestones. On this basis, KARAMATA *et al.* (1999, 2000a) distingushed the eastern and western branches of the Vardar Zone, which are separated by the almost north-south stretching Kopaonik unit. They regarded the eastern branch as a relict of the main Vardar Ocean, which was closed in the Upper Jurassic, and the western one as a younger oceanic realm, closed in the Upper Cretaceous.

In this paper, the K/Ar radiometric ages of the Ždraljica ophiolitic complex (hereafter ŽOC) are presented and discussed. The complex is located near Kragujevac in central Serbia, about 150 km south of Belgrade and it covers about 30 km^2 (Fig. 1). This ophiolite complex is geotectonically situated along the easternmost margin of the Vardar Zone and shares the following general characteristics of the ophiolites of the eastern branch of the Vardar zone (KARAMATA *et al.*, 1999): (i) they are predominantly composed of igneous members of ophiolites, (ii) they are characterized by rare exposures of peridotitic bodies commonly severely serpentinized, (iii) they contain various granitic rocks, and (iii) their overstep sequence is Upper Jurassic in age (e.g. KARAMATA *et al.*, 2005).

Analytical procedure

The radiometric K/Ar data was obtained using a magnetic sector mass spectrometer designed in the Institute for Nuclear Research ATOMKI of the Hungarian Academy of Sciences in Debrecen (Hungary). The analytical procedure was given in detail by BALOGH (1985).

The instrument works in the static mode with a radius 150 mm and a deviation of 90°. For the Ar-analysis, the line for the Ar-extraction was also used. The most suitable samples were crushed, care being taken that the smallest grain size was not less than 0.05 mm. Dusty material was avoided because it increases the binding of atmospheric argon to the particle surfaces, which leads to an increase of the Ar-content of the samples. For the whole rock analyses, the samples were only roughly crushed, but for mineral analyses separation by firstly an electromagnetic separator and subsequently by heavy liquids was performed. In the final stage, the most fresh grains were picked out under binocular lenses.

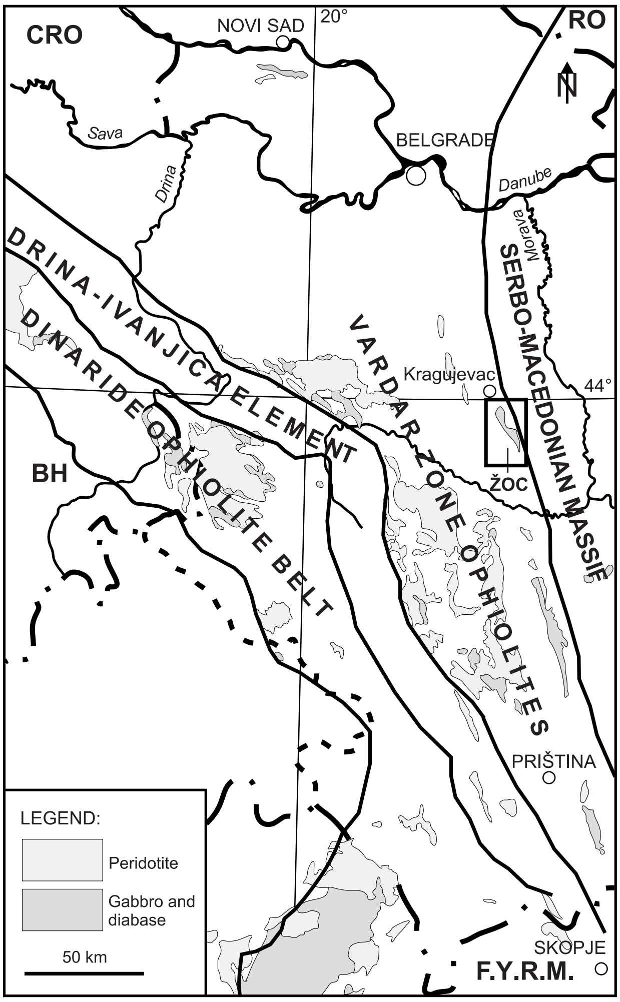

Fig. 1. Geotectonic position of the Ždraljica ophiolitic complex (ŽOC). The terrane boundaries and distribution of Mesozoic ophiolites are given by KARAMATA *et al.* (1994) and DIMITRIJEVIĆ (1992).

Petrography and Geochemistry

Igneous rocks of the ŽOC can be divided into two petrogenetically different rock suites: (1) a predominant suite of tholeiitic affinity and (2) a subordinate suite of a calc-alkaline affinity (RESIMIĆ, 2000).

Tholeiitic affinity suite

The tholeiitic suite is represented by massive gabbros and diabases with subordinate cumulitic gabbros and peridotites. Basalts and plagiogranites are very rare. The whole suite was metamorphosed under low- to medium-T conditions. Therefore, some rocks can be classified as spilites, metadiabases or even greenschists.

Although the ŽOC is a dismembered complex, an almost complete oceanic crust section can be reconstructed. The deepest part of the ŽOC is represented by highly serpentinized cumulitic wherlites, which developed as rare pods and layers within gabbros. They are allotriomorphic and orthocummulitic in texture, composed of olivine ($Fo_{85.73}$) and diopside (Mg# 89.8). Gabbros appear as irregular blocks or sheets of massive and cummulitic facies. Texturally, they are coarse-, rarely fine-grained and poikilitic to coarse-grained ophitic and orthocummulitic, and contain plagioclase (fresh: 63.8–97.6 % An or albitized: 3.7–19.6 % An), clinopyroxene (diopside with Mg# 73.0–93.3 and augite with Mg# 62.4–86.7) and, very rarely, totally serpentinized olivine. The accessories are magnetite, sphene and apatite. Secondary minerals are epidote, uralite, chlorite, tremolite, calcite, serpentine, pyrite and, rarely magnesiohornblende. Diabases appear as single dykes or dyke-swarms intruding gabbros or irregular massive bodies. They show ophitic, intergranular or intersertal textures. The main mineral phases are plagioclase (70.2–85.7 % An) and augite (Mg# 61.3–80.2), while accessories are magnetite, sphene and altered glass. Metamorphosed diabases are mostly composed of albite (2.3–6.9 % An), epidote/zoisite, uralite, tremolite, magnesiohornblende, calcite and pyrite. Basalts mainly occur as pillow lavas and, primary and redeposited, hyalloclastites. They have glomeroporphyritic, ophitic, aphyric and intersertal textures. Phenocrysts are albitized plagioclase (7.7–11.7 %An), relicts of augite (Mg# 60.2 and 77.7), opaques and serpentinized olivine, while the groundmass is composed of chloritised glass and microlites of plagioclase. Plagiogranites occur as small dyke- or sill-like intrusions and irregular bodies within diabases and fine-grained gabbros. They display hypidiomorphic and fine-grained granular texture. They are composed by albite and quartz, accessories are magnesiohornblende, magnetite and apatite, while epidote/zoisite and chlorite are secondary products.

The geochemical characterization of these suite is given in detail in Resimić (2000) and Resimić-Šarić *et al.* (2004) and will only be summarized here. According to their major element composition, these rocks show a tholeiitic affinity (Irvine & Baragar, 1971). Their MORB characteristics are suggested by HFSE ratios, e.g. Zr/Y(around 2.5) and Ti/Y (around 240) (Saunders & Turney, 1984). On Nb*2–Zr/4–Y (Meshede, 1986) and V vs. Ti (Shervais, 1982) diagrams these rocks occupy both MORB and VAB fields. This is confirmed by the chemical compositions of clinopyroxenes plotted on Ti+Cr vs. Ca (Letterier *et al.*, 1982) and F_1 vs. F_2 (Nisbet & Pearce, 1977) diagrams. In addition, the plagiogranites show an ORG affinity according to (Y+Nb) vs. Rb and Y vs. Nb (Pearce *et al.*, 1984) discrimination diagrams. Geochemical modelling shows that the most primitive rocks of the tholeiitic suite could have originated from 10–15 % non-modal batch melting of a spinel-lherzolite ($Ol_{57}Opx_{25.5}Cpx_{15}Sp_{2.5}$) source. The most important process of magma modification was fractional crystallization (F=0.6), with the fractionating assemblage: $Pl_{52.9}Cpx_{12.5}Ol_{26.1}Ttn_{2.9}Ap_{4.4}Mgt_{1.0}$.

Suite of calc-alkaline affinity

The calc-alkaline affinity suite is predominantly represented by leucocratic granodiorites and granites and by rare quartzdiorites and quartzmonzodiorites. These rocks have a similar mode of occurrence. They generally apear as irregular bodies or smaller dykes, which show sharp and sheared boundaries in contact with gabbros.

Leucocratic granodiorites and granites are hypidiomorphic medium- to coarse-grained in texture. Poikilitic, perthitic and granophiric textures are also observed. They consist of quartz, K-feldspar, albite (0–6.9 % An), chloritised biotite and amphibole. Accessory phases are apatite, magnetite, and zircon, while sericite and chlorite are alteration products. Quartzdiorites and quartzmonzodiorites show a medium-grained, hypidiomorphic texture. The main minerals are plagioclase (48.1–42.2 % An) and magnesiohornblende (Mg# 0.63–0.74), while K-feldspar is very rare and occurs only as interstices in quartzmonzodiorites. The accesorites are sphene, apatite, zircon and opaque minerals.

Geochemically, both the leucocratic granodiorites/granites and the quartzdiorites/quartzmonzodiorites are calc-alkaline and differ from plagiogranites in having higher LILE and lower HFSE and Cr, Ni contents. However, the leucocratic granodiorites/granites are peraluminous and have a similar composition to the Furka and Fanos granitoids found more to the south in the F.Y.R. of Macedonia and Greece, respectively. These granitoid rocks occur within the ophiolitic complexes which geotectonically belong to the eastern branch of the VZ. In addition, the ŽOC leucocratic granitoids show syn- to postcollisional character (Harris *et al.*, 1986, Pearce *et al.*, 1984), as do the Furka and Fanos granitoids (Šoptrajanova, 1967, Christofides *et al.*, 1990). On the other hand, the ŽOC quartzdiorites and quartzmonzodiorites are metaluminuous and are akin to VA-granitoids, i.e. they show a pre-collisional character on the above mentioned discrimination diagrams.

K/Ar data of the ŽOC

The K/Ar radiometric age determinations of the whole rock (w.r.) samples and the mineral separates (amphibole, plagioclase and K-feldspar) from both the tholeiitic and calc-alkaline suites of the ŽOC are presented in Table 1 and Fig. 2. There is a wide range of age data (168.4–77.3 Ma), which probably mark various events in the evolution of the ŽOC. Thus, these radiometric data can be interpreted as showing: (a) the age of igneous processes and (b) the time of various metamorphic events.

Table 1. K/Ar radiometric data for minerals and rocks of the ŽOC.

Rock type	Sample number	Analised material	K (%)	$Ar_{(rad)}$ (cm^3/g)	$Ar_{(rad)}$ (%)	Age (Ma ± σ)
gabbro	8-B	amphibole	0.0765	4.718×10^{-7}	26.8	152.0 ± 21.0
		plagioclase	0.32	1.250×10^{-6}	19.5	97.8 ± 7.3
diabase	E-550	w.r.	0.285	1.567×10^{-6}	58.7	136.2 ± 7.7
		plagioclase	0.71	3.066×10^{-6}	68.3	107.8 ± 4.5
quartzdiorite	V-306/7	amphibole	0.38	2.607×10^{-6}	62.8	168.4 ± 6.7
		plagioclase	0.75	2.683×10^{-6}	54.0	90.0 ± 3.7
quartzmonco-diorite	V-400a	feldspars	2,74	1.233×10^{-5}	83.1	112.3 ± 4.3
		amphibole	1.30	6.725×10^{-6}	59.1	128.5 ± 5.2
	V-353c	feldspars	2.416	1.172×10^{-5}	68.5	120.7 ± 4.8
		amphibole	1.249	7.340×10^{-6}	78.9	145.2 ± 5.7
leucocratic granite	V-306/6	w.r.	4.60	1.819×10^{-5}	94.0	99.0 ± 3.8
		K-feldspar	7.45	2.286×10^{-5}	89.0	77.3 ± 3.6
	V-353b	plagioclase	1.55	5.917×10^{-6}	79.0	95.6 ± 3.8

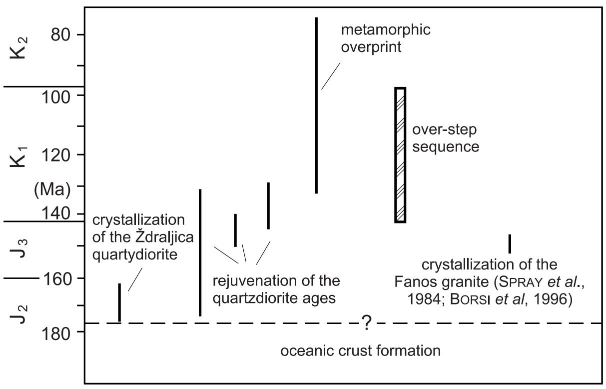

Fig. 2. K/Ar data for analysed minerals and rocks of the ŽOC.

The oldest ages were obtained on amphiboles from the quartzdiorite samples. The value of 168.4±6.7 Ma (sample V-306/7) most likely represents the age of intrusion of the quartzdiorite magma. The effects of certain rejuvenation can be taken into consideration because partial chloritization of the amphibole from the quartzmonzodiorites could have caused some loss of radiogenic Ar. This can explain the younger estimate of 145.2±5.7 Ma for the amphibole from sample V-353c.

Amphiboles from a gabbro sample (8-B) revealed an age of 152 Ma but with a very high error of ±21 Ma. It is not likely that the crystallization age of these rocks is lower than 152 Ma because this gabbro is intruded by the above mentioned quartzdiorites. Therefore, this estimate indicates some metamorphic overprints and may represent the age of the emplacement of the Ždraljica ophiolites. The much younger whole rock age of 136±7.7 Ma obtained on a whole-rock diabase sample for certain post-dates the age of ophiolite emplacement.

The ages of the feldspars from all lithologies of the ŽOC revealed a similar range of 120.7–77 Ma. These ages suggest that the whole ŽOC underwent a metamorphic overprint during the Cretaceous. Of the dated minerals, K-feldspar loses Ar at the lowest temperature (130±15° C) and this is in accordance with the youngest ages measured on the K-feldspar from the leucocratic granite.

Discussion and concluding remarks

The ophiolites of Ždraljica predominantly consist of tholeiitic MORB-affinity igneous rocks, which represent slices of the ancient oceanic crust. The crystallization age of these rocks could not be directly radiometrically dated by the K/Ar method. All the K/Ar estimates on tholeiitic rocks are younger than the presumed emplacement age of the ophiolites. The time of emplacement can be inferred from the fact that the ŽOC is associated with a Middle/Upper Jurassic diabase-chert formation (MARKOVIĆ *et al*., 1968, DOLIĆ *et al*., 1981) and that its immediate overstep sequence is represented by Tithonian limestones and Lower Cretaceous paraflysch (MARKOVIĆ *et al*., 1968; DOLIĆ *et al*., 1981).

The magmatic age of the ŽOC can possibly be inferred on the basis of the crystallization age of the calc-alkaline quartzdiorites, which was estimated at around 168 Ma. These rare rocks are metaluminous and show a pre-collisional VA-character. This can indicate that at least some parts of the ŽOC are relicts of an immature island arc. In this context, the minimal formation age of this oceanic crust can be regarded as Middle/Late Jurassic. On the other hand, the crystallization age of the leucocratic granitoids cannot be inferred from the available K/Ar data. Their geochemical characteristics suggest that they formed under a syn- to post-collisional setting, i.e. after the emplacement of the ŽOC. As mentioned above, petrochemically similar counterparts are the Furka and Fanos granitoids which occur more to the south, within the Guevgeli ophiolitic complex. The Fanos granite was dated 148±3 Ma (K/Ar on biotite; SPRAY *et al*., 1984) and 150±2 Ma (K/Ar and Rb/Sr on biotite; BORSI *et al*., 1966). These ages can correspond to the time of intrusion of Ždraljica leucocratic granitoids in the syn- to post-collisional phase following the emplacement of ophiolites.

The above presented data concerning the Ždraljica ophiolite complex support the interpretation based on the existence of two-branches within the Vardar Zone. In many geotectonic interpretations, the ophiolites of the Vardar Zone were generally believed to be related with Jurasssic igneous sequences situated more to the north in the South Apuseni (BLEAHU *et al*., 1981;

NICOLAE, 1995; BORTOLOTTI *et al*., 2002, and references therein). They are mainly based on K/Ar Upper Jurassic ages available for south Apuseni (Mures) ophiolites (167.8±5 Ma – NICOLAE *et al*., 1992). In addition, south Apuseni ophiolites are associated with radiolarian cherts of Callovian to Oxfordian age (LUPU *et al*., 1995). However, these interpretations did not take into account that only the eastern part of the Vardar Zone ophiolites can be considered as a continuation of these occurrences. In this context, important evidence is provided by this study. For instance, the following characteristic of the ŽOC are similar to the south Apuseni ophiolites: (i) the Ždraljica ophiolitic segment is dominated by igneous members with only subordinate peridotites, (ii) it was formed as a MORB-type oceanic crust in Middle Jurassic, (iii) it contains calc-alkaline rocks of a VA-signature, which presumably originated in a pre-collisonal setting, (iv) the whole ophiolitic unit was intruded by leucocratic peraluminous granites after the emplacement, and (v) its overstep sequence is represented by Titonian limestones and Lower Cretaceous flysch. The southern continuation of the eastern branch of the Vardar Zone are the Kuršumlija (south Serbia) and Guevgeli (F.Y.R.M. and northern Greece) ophiolites. The Kuršumlija ophiolites are situated only about 150 km southwards from the ŽOC. They are very similar to Ždraljica rocks with respect to the characteristics of the facies and petrochemistry (RESIMIĆ-ŠARIĆ, unpublished) but radiometric data are still not available for these rocks. On the other hand, the Guevgeli ophiolites also meet the above summarized characteristics (e.g. IVANOVSKI, 1970). It is worth noting that the specific geotectonic position of the Guevgeli ophiolites with respect to more westwardly situated Vardar ophiolites was emphasized in the seventies by MERCIER (1973).

The presented data set is not sufficient to explain the age and formation of the Ždraljica ophiolites in detail. However, there is evidence that this is a specific Jurassic ophiolitic section with possible dismembered counterparts in the north and in the south. They probably represented (an) oceanic domain(s) situated along the southern European margin during the Jurassic. After the closure and accretion, they probably formed as a unique ophiolite belt. The southern part of the belt more or less preserved its continuation, while the northern parts came into the present position during the tectonic escape of Adria-related microplates during the Late Paleogene–Early Neogene.

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Резиме

Радиометријске K/Ar старости као доказ геодинамичке еволуције Ждраљичког офиолитског комплекса (централна Србија)

Офиолитски комплекс Ждраљице (ЖОК) налази се надомак Крагујевца, око 150 km јужно од Београда. Геотектонски, он припада главном (источном) ободу Вардарске зоне.

Магматске стене ЖОК-а могу се поделити на свиту толеитског афинитета, која доминира, и свиту калко–алкалних карактеристика. Толеитска свита представљена је масивним габровима и дијабазима, подређено кумулатним габровима и перидотитима, док су базалти и плагиограniти веома ретки. Ове стене показују MORB-карактеристике према садржајима HFS елемената, нпр. Zr/Y(~ 2.5) и Ti/Y (~ 240). На дијаграмима Nb*2–Zr/4–Y и V vs. Ти заузимају поља и MORB-а и VAB-а, што је потврђено и хемијским саставом пироксена на дијаграмима Ti+Cr vs. Ca и F_1 vs. F_2. Плагиограниti

показују ORG афинитет. Калко-алкална свита представљена је претежно леукократним гранитима и гранодиоритима и ретким кварцдиоритима и кварцмонцодиоритима. Према геохемијским одликама ове стене се разликују од плагиогранита по већем садржају LILE и мањем HFSE, Cr и Ni. Леукократни гранити и гранодиорити су пералуминијски и сличног су састава као гранити Фурке (Македонија) и Фаноса (Грчка). Ову сличност потврђује и њихов син- до пост-колизиони карактер. Са друге стране, кварцдиорити и кварцмонцодиорити су метаалуминијски, блиски VA гранитоидима и показују пре-колизиони карактер.

Старост кристализације толеитских стена MORB афинитета није директно одређена K/Ar мерењима. Све добијене старости су млађе од претпостављеног времена смештаја ЖОК-а које је одређено на основу чињенице да је ЖОК асоциран са средњо/горњојурском дијабаз-рожначком формацијом и да overstep секвенцу представљају титонски кречњаци и доњокредни парафлиш.

Старост магматских процеса од око 168 Ма указује кристализацију калко-алкалног преколизионог и метаалуминијског кварцдиорита VA-афинитета. Ово указује да су барем неки делови ЖОК-а реликти незрелог острвског лука, односно, да се као минимална старост образовања океанске коре може сматрати средња/горња јура. С друге стране, време кристализације леукократних гранитоида није се могла добити радиометријским K/Ar датовањем. Њихове одлике, међутим, указју да су образовани у син- до постколизионим условима, односно, после смештаја ЖОК-а, слично гранитима Фурке и Фаноса. Старост гранита Фаноса износи 148 ± 3 Ма (K/Ar на биотиту и 150 ± 2 Ма (K/Ar и Rb/Sr на биотиту). Ове старости могле би да одговарају и времену интрудовања Ждраљичких леукократних гранитоида у син- до пост-колизионој фази која је пратила смештај офиолита.

Приказани подаци о ЖОК-у потврђују интерпретацију о постојању два огранка унутар Вардарске зоне – источни или главни за који се сматра да је затворен током јуре и западни који је затворен у креди. Наиме, ЖОК има одлике које су карактеристичне и за друге офиолитске комплексе унутар источног обода Вардарске зоне. У многим геотектонским интерпретацијама офиолити јужних Апусена сматрају се делом јурске магматске секвеце Вардарске зоне, углавном на основу K/Ar горњојурских (167.8 ± 5 Ма). Поред тога ови офиолити су асоцирани са радиоларитима каловијске до оксфордске старости. Ове интерпретације, међутим, нису узимале у обзир да се само у источном ободу Вардарске зоне налазе појаве офиолита које представљају континуитет. Следеће карактеристике ЖОК-а су сличне са одликама офиолита јужних Апусена: (а) комплекс је изграђен углавном од магматских чланова офиолита, (б) океанска кора MORB-типа образована је у средњој јури, (в) комплекс садржи калкоалкалне стене VA афинитета, које су вероватно преколизионог карактера, (г) цела офиолитска секвенца је после смештаја интрудована леукократним пералуминозним гранитоидима и (д) overstep секвенца је представљена титонским кречњацима и доњокредним флишом. Наставак источног обода Вардарске зоне на југ представљају офиолити Куршумлије и Ђевђелије.

Приказани сет података није довољан да се детаљно објасне старост и настанак ЖОК-а. Међутим, доказано је да је реч о специфичном јурском офиолиту који се на север и југ наставља са сличним појавама. Сви они вероватно представљају један или више јурских океанских домена дуж јужне европске маргине, који су после затварања и акреције образовали јединствен офиолитски појас. Јужни део овог појаса сачувао је мање-више свој континуитет, док је његов северни део дошао у данашњи положај за време горњег палеогена–доњег неогена.