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VERMICULITE DEPOSITS IN THE BALKAN PENINSULA

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ABSTRACT

Although vermiculite or mica - vermiculite intercalations are common, large deposits of economic interest are rare due to their formation conditions, which require a complex combination of endogenic and exogenic processes, in a very definite succession and during a long period of time. Recently, vermiculite deposits have been located in the Balkan peninsula, the most significant of them occurring in the Srednogorie region, the Rhodope and Serbomacedonian massifs. Several small occurrences were also found at Zidani, Vavdos, Gerakini, Valandovo - Rabrovo and elsewhere.

Following the scheme proposed by Lvova (1974), which is based on the associated rocks and the origin of vermiculite, the majority of the Balkan peninsula deposits can be classified to Group B. Furthermore, they can be distinguished into to the types 3 and 4. Type 3 is associated with tectonites, which are highly or completely serpentinized and includes vermiculite - hydrophlogopite deposits. Since parent mica is a high-Mg phlogopite the main components, which are vermiculite and hydrophlogopite are characterized by f_{com} ranging between 4 and 12. Type 4 is associated with the magmatic (cumulate) sequence of ophiolite complexes (peridotites, pyroxenites and gabbros). The main component is hydrophlogopite. Parent mica, which is phlogopite has a higher Fe content compared to type 3, and the main ore components are characterized by a high f_{com} ranging from 15 to 30. Thus, both types differ in their f_{com} resulting difference in the degree of hydration, physical and technical properties of vermiculites.

Structural and petrological data of the Balkan peninsula are not favor for hosting vermiculite deposits associated with alkaline - ultramafic and carbonatite complexes, belonging to Group A. However, the large deposits recently discovered have natural properties of acceptable quality and are of economic interest. Therefore, the Balkan peninsula is considered as a potential target for vermiculite prospecting and and further exploration works.

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ΣΥΝΟΨΗ

Αν και η εμφάνιση βερμικουλίτη ή της παραγένεσης μαρμαρυγία - βερμικουλίτη είναι συνήθης, αξιόλογα από οικονομική άποψη κοιτάσματα βερμικουλίτη, είναι σπάνια. Αυτό αποδίδεται στις συνθήκες σχηματισμού του βερμικουλίτη που απαιτούν ένα περίπλοκο συνδυασμό ενδογενών και εξωγενών διεργασιών, με μία καθορισμένη διαδοχή κατά την διάρκεια μεγάλης χρονικής περιόδου. Πρόσφατα, εντοπισθηκαν κοιτάσματα βερμικουλίτη στην Βαλκανική χερσόνησο. Τα σημαντικώτερα γνωστά κοιτάσματα ευρίσκονται στις περιοχές Srednogorie, την Ροδοπική και την Σερβομακεδονική μαζα. Επίσης, αρκετές μικρές εμφανίσεις βερμικουλίτη έχουν εντοπισθεί στις περιοχές Ζιδανίου, Βάβδου, Γερακινής, Valandovo - Rabrovo και αλλού.

Με βάση τα πετρώματα που φιλοξενούν τα κοιτάσματα ή εμφανίσεις βερμικουλίτη και τις συνθήκες σχηματισμού τους, η πλεινότητα αυτών ταξινομείται στην ομάδα Β -σύμφωνα με το σχήμα που προτάθηκε από την Lvova (1974) - και διακρίνονται στούς τύπους 3 και 4. Ο τύπος 3 συνδέεται με τεκτονίτες (περιδοτίτες της μανδυακής σειράς), οι οποίοι είναι εντόνως ή πλήρως σερπεντινιωμένοι και περιλαμβάνουν κοιτάσματα βερμικουλίτη - υδροβιοτίτη. Τόσο ο φλογοπίτης που αποτελεί το μητρικό ορυκτό όσο και τα κύρια ορυκτά βερμικουλίτης και υδροβιοτίτης είναι Mg-πλούσια (f_{com} = 4 - 12). Ο τύπος 4 συνδέεται με την μαγματική (σωρειτική) σειρά των οφιολιθικών συμπλεγμάτων (περιδοτίτες, πυροξενίτες και γάββρους). Το κύριο συστατικό είναι υδροφλογοπίτης. Ο φλογοπίτης που αποτελεί το μητρικό ορυκτό είναι πλουσιώτερος σε σίδηρο, σε σχέση με τον τύπο 3 και τα κύρια ορυκτά του μεταλλεύματος χαρακτηρίζονται επίσης από υψηλές τιμές του δείκτη f_{com} (15 - 30). Συνεπώς, οι δύο τύποι διαφέρουν ως προς τον συντελεστή f_{com}, ο οποίος συνεπάγεται διαφορετικό βαθμό διόγκωσης και φυσικο - τεχνικών ιδιοτήτων του βερμικουλίτη.

Τα γεωλογικά και πετρολογικά χαρακτηριστικά της Βαλκανικής χερσονήσου δεν παρέχουν ενδείξεις για την παρουσία της ομάδας Α βερμικουλίτη. Ωστόσο, δεν υπάρχει αμφιβολία ότι στην Βαλκανική χερσόνησο φιλοξενούνται μεγάλα κοιτάσματα βερμικουλίτη με τα απαιτούμενα χαρακτηριστικά αποδεκτής ποιότητας. Κατά συνέπεια, φαίνεται ότι η Βαλκανική χερσόνησος είναι ενδιαφέρουσα για περαιτέρω έρευνα για εντοπισμό και εκμετάλλευση βερμικουλίτη.

INTRODUCTION

Vermiculite, which is an industrial mineral, was recently found in the Balkan peninsula. The name vermiculite is generally applied to a group of hydrated ferro-magnesian minerals including vermiculite itself, and intercalations of phlogopite-vermiculite, which are characterized by their ability to expand under heating. The so-called exfoliation, produces a lightweight material, characterized by large cation exchange capasity, organic complexing ability, water holding capasity, heat and acustic insulation. Due to these properties, vermiculite is used in construction, agriculture, fireproof, chemical and general industry.

Although vermiculite and phlogopite-vermiculite intercalations is a quite common mineral association, large deposits of economic interest are rare, due to their formation conditions, which require a complex combination of endogeneous and exogeneous processes, in a very definite succession and during a long period of time.

Detailed descriptions of the geology, structure, petrology, mineralogical and chemical composition of vermiculite deposits and occurrences in the Balkan peninsula, have been given by previous investigators (Barska, 1958, 1972a,b; Skarpelis and Dabitzias, 1987; Zhelyaskova - Panayotova, 1989a,b,c; Dabitzias and Perdikatsis, 1990; Zhelyaskova - Panayotova et al., 1992). For the time being the most significant vermiculite deposits were located in the Srednogorie region, the Rhodope and Serbomacedonian massifs. Several small vermiculite occurrences were also found at Zidani, Valandovo - Rabrovo and elsewhere.

Market conditions, location, size, grade and quality are some factors affecting the commercial value and development of a deposit. Recently, research projects in Bulgaria and Greece are dealing with the estimation of vermiculite potential and the problem of its origin. Particularly, detailed investigations have been made on the Ichtiman region, Bulgaria and Ascos, Greece (Figs 1 & 2). In the present paper, on the basis of the available data a classification of the vermiculite deposits in the Balkan peninsula is attempted. Also, in order to determine the age of micatization process, some K-Ar isotopic data are given.

CHARACTERISTICS OF VERMICULITE DEPOSITS AND OCCURRENCES, AND THEIR CLASSIFICATION

Among the attemps which have been made to classify vermiculite deposits is that proposed by Lvova (1974), based on the associated rocks and the geodynamic conditions of their formation. It has been accepted that the characteristic features of the parent mica such as structure, f_{com}*, F quantity e.t.c, which depend on the host rock, are the main controlling factor of the vermiculite deposits. Thus, endogeneous processes, which are reflected on the character and degree of parent mica are considered to be among the main factors controlling vermiculite deposits, although vermiculite is formed under the influence of exogeneous processes, such as weathering. The larger commercial vermiculite deposits are usually found to be associated with weathering crusts of sialilic type. Their middle zone is the best developed one, whereas the lower part usually is complicated due to tectonic events. Their thickness varies from a couple to several tens of meters. The intensity of the weathering process is affected by the climatic conditions and the duration of the weathering crust formation. Also, exchange cation reactions may be affected by the composition of the underground water.

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* $f_{com} = (2Fe_2O_3 + FeO).(2Fe_2O_3 + FeO + MgO)^{-1} . 100$

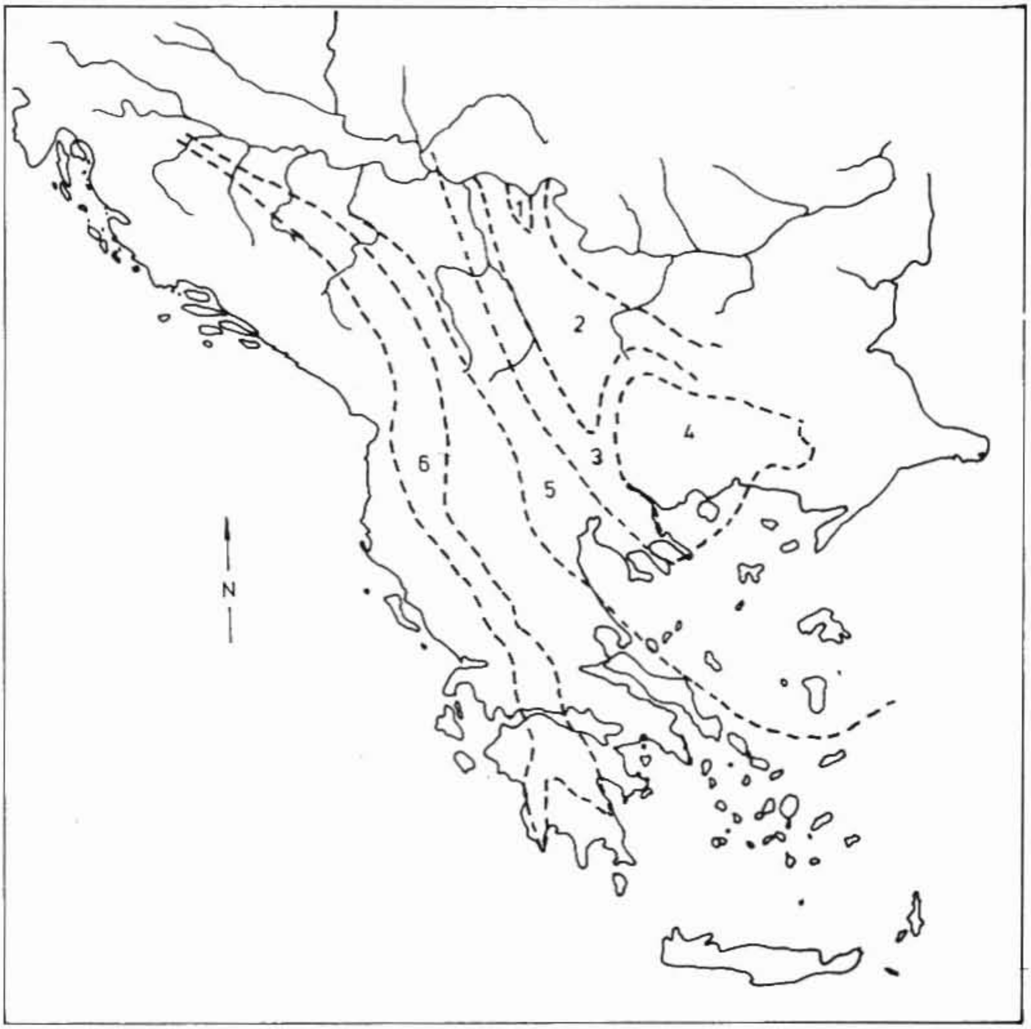

Fig. 1. The ophiolite belts of the Balkan peninsula (after Karamata et al., 1989). Symbols 1: metaophiolites in Precambrian basement; 2: metaophiolites in the Caledonian basement of the Carpatho-Balkan system and Serbo-Macedonian massif; 3: ophiolites in the Serbo-Macedonian massif; 4: ophiolites and metaophiolites of the Rhodope massif, 5: ophiolites of the Vardar zone; 6: ophiolites in the Dinarides, Albanides and Hellenides

Εικ. 1. Οι οφιολιθικές σειρές της Βαλκανικής Χερσονήσου (Κατά Καραμάτα κ.α., 1989). Σύμβολα 1: μεταοφιόλιθοι στο υπόβαθρο Προκαμβρίου ηλικίας, 2: μεταοφιόλιθοι στο υπόβαθρο Καληδονίου ηλικίας του Καρπαθο-Βαλκανικού συστήματος και της Σερβο-Μακεδονικής μάζας, 3: οφιόλιθοι της Σερβο-Μακεδονικής μάζας, 4: οφιόλιθοι και μεταοφιόλιθοι της Ροδοπικής μάζας, 5: οφιόλιθοι της ζώνης Αξιού, 6: οφιόλιθοι των Δειναρίδων, Αλβανίδων και Ελληνίδων

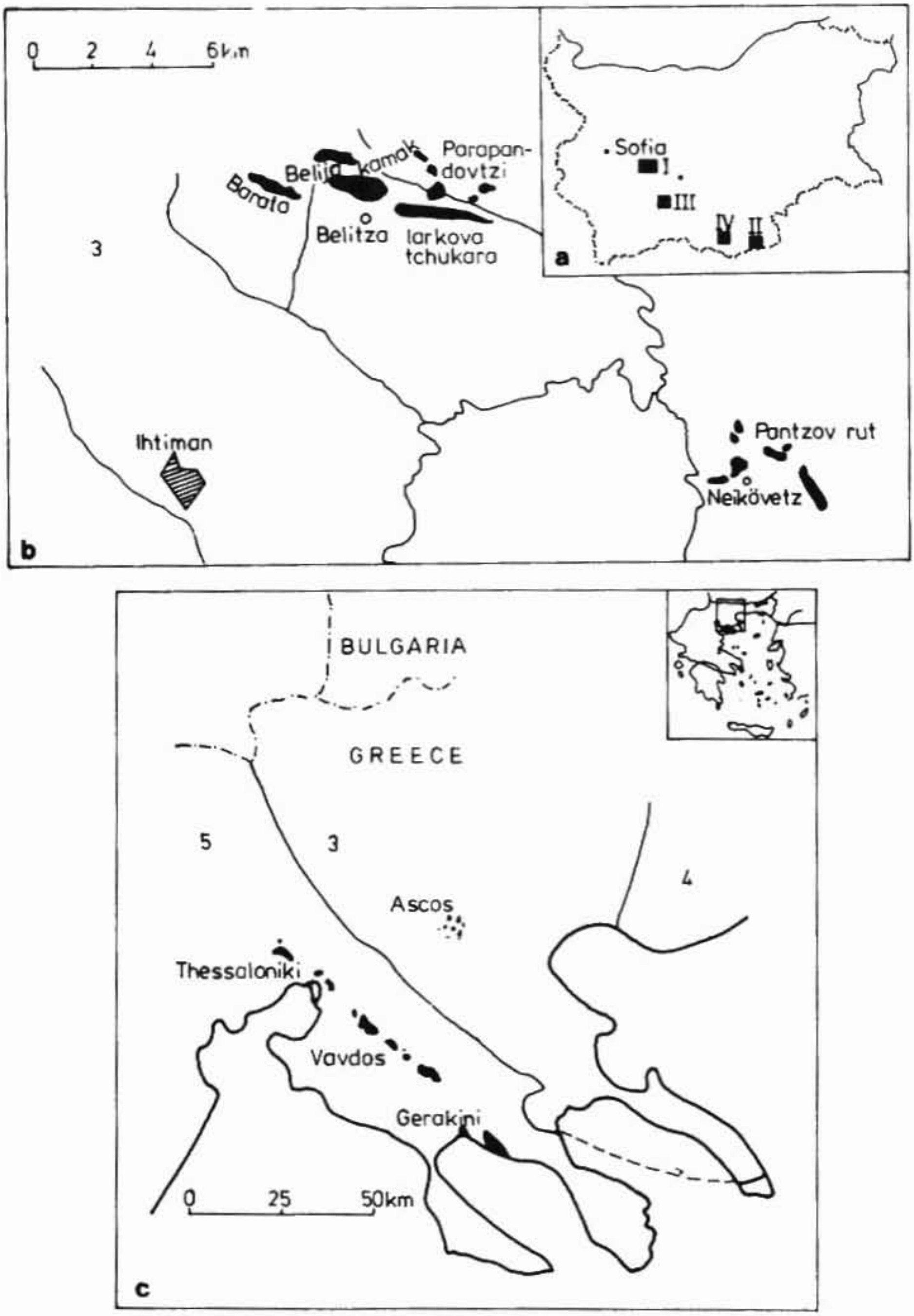

Fig. 2. Vermiculite deposits and associated ophiolites (a) in Bulgaria - I: Ihtiman; II: Avren; III: Velingrad and IV: Ardino, (b) in the Ihtiman region and (c) in the Serbo-Macedonian massif.
Εικ. 2. Κοιτάσματα βερμικουλίτη και οφιολιθικά συμπλέγματα που συνδέονται με αυτά (a) στην Βουλγαρία (b) στην περιοχή Ihtiman και (c) στην Σερβο-Μακεδονική μάζα.

According to the classification proposed by Lvova (1974) four groups and nine types of vermiculite deposits can be distinguished: **GROUP A, TYPE 1 & 2**: the most significant industrial vermiculite types are included, such as the vermiculite - phlogopite type deposits, associated with alkaline - ultrabasic and carbonatite complexes. Examples of this type include the Phalaborwa deposit in RSA, which characterized by both, the high quality and large vermiculite reserves. The Kovdor deposit in Russia and Libby deposit, Montana in USA, belong this type too (Lvova, 1974).

Based on the same classification described above with slight modification, the vermiculite deposits and occurrences found in the Balkan peninsula, can be classified into **GROUP B**, and more specifically they can be distinguished into the types 3 and 4 (Table 1). Type 3 is associated with tectonites, and type 4 with the magmatic (cumulate) sequences of ophiolite complexes, and sometimes they are found in a close spatial association, especially in the Ichtiman region. They occur as ophiolitic boudins within the highly crystaline basement (consisting of gneisses, amphibolites, mica - schists and marbles), which have been affected by metamorphism into amphibolite and amphibolite - epidote facies (Karamata et al., 1989).

The vermiculite deposits and occurrences of the Balkan peninsula are distributed in the Ichtiman region (Barata, Beliya - Kamak, Parapandovtsi, Yarkova chukara, Pantsov rut and Neikovets), Avren region, chubrika in the Ardino area, Dorkovo and Kostandovo in the Velingrad area, and elsewhere (Bulgaria), Ascos, Vavdos, Gerakini Zidani (Greece), Valandovo - Rabrovo (former Yugoslavia). They were found in a spatial and genetic association with various types of ferromagnesian rocks, which have been affected by a strong micatization.

TYPE 3, includes vermiculite - hydrophlogopite deposits, usually associated with tectonites (mainly harzburgites), which are highly or completely serpentinized. Amphiboles (tremolite, actinolite), chlorite, talc and carbonate minerals are also present. Although the main component is vermiculite, hydrophlogopite with a varying proportion of vermiculite are also present in minor quantities at the lower parts of the profils (the term hydrophlogopite is used not only for the proportion 1:1, but for all interlayers of phlogopite - vermiculite). Micatization is widespread over the smaller boudins and within larger bodies. Since, parent mica is a high - magnesian phlogopite, with f_{com} = 4 - 12, both vermiculite and hydrophlogopite are high - magnesium too (f_{com} = 4 - 10). Appreciable substitution of K cations by Mg^{2+} is common. The most common mineralization types are in the form of massive, stockwork, veins along cracks, pockets or lenticular zones at the contacts between ultrabasic rocks and other rocks. The thickness of the vermiculite - bearing outcrops is variable ranging from a few to 30 meters. The mineralization along the contacts between ultrabasic rocks and pegmatites is better developed at the areas of Yakovitca, Golyamo Kamenyane, Stanchovitsa, Parapandovtsi and elsewhere.

The Parapandovtsi deposit (Ichtiman region) is a typical example of type 3, with bulk material containing average vermiculite 16 % , and the ore reserves are 850.000 tons. Type 3,

Table 1. Classification of vermiculite deposits and occurrences in the Balkan peninsula

Πίνακας 1. Ταξινόμηση κοιτασμάτων και εμφανίσεων βερμικούλίτη στην Βαλκανική Χερσόνησο

GROUP B

1. **Type 3**, associated with tectonites (harzbourgites)
Vermiculite - hydrophlogopite (f_{com} = 4-10) deposits
parent mica : high - Mg phlogopite (f_{com} = 4-12)
mineralization type: massive, stockwork, veins, irregular masses

Typical examples:
Ichtiman region
- Parapandovtsi deposit
- Pantsov rut
- Divyaka

Rhodope massif
- Seltce
- Choubrika
- Golyamo Kamenyane

2. **Type 4**, associated with the cumulate sequence
Hydrophlogopite (hydrobiotite) - vermiculite (f_{com} = 15-30) deposits
parent mica = phlogopite - Fe-phlogopite (f_{com} = 20 - 40)
mineralization type: massive, stockwork, veins, irregular masses

Typical examples:
Ichtiman region
- Beliya - kamak
- Barata and Jarkova Choukara

Rhodope massif
- Avren deposits

Serbo-Macedonian massif
- Ascos, Vavdos, Gerakini

GROUP C

3. **Type 5** Small vermiculite occurrences associated with magnesian skarns

Typical examples:
- Malko Turnovo area
- Kroumovo,

GROUP D

Type 9 Hydrobiotite occurrences associated with biotite gneisses and schists

Typical example : Dolen area, W. Rhodope massif also includes the Pantsov rut and Divyaka deposits, in the same region and a number of smaller deposits of the Rhodope massif (Seltce, Chubrika and Golyamo Kamenyane). Those deposits seem to be comparable with the Young River (Australia), Buldimscoe and Andreevscoe (Russia) (Lvova, 1974).

TYPE 4, includes hydrophlogopite (hydrobiotite) - vermiculite, associated with the magmatic sequence of ophiolite complexes. They are represented by peridotite - pyroxenite (mostly diopsidite) - gabbro. Host rocks with variable degree of serpentinization and amphibolization (tremolite - actinolite), have been slightly affected by talcitization and are often intruded by pegmatites. It is noticeable, that vermiculite - bearing rocks from the Ichtiman region exhibit an enrichment in rare earth elements and apatite (up to 12%). In the Ascos area, Serbomacedonian massif, vermiculite irregular masses with tremolite + chlorite, and talc with disseminated chlorite and serpentinite pockets, form in that order descrete zones, at the contact between gneiss and serpentinite. Those zones exhibit sharp contacts and variable thickness, varying from a few tens of centimeters up to some 5 meters (Dabitzias and Perdikatsis, 1990). The zone of chlorite schists contains coarce grains of magnetite.

The main ore component is hydrophlogopite (hydrobiotite). Vermiculite occurs only at the uppermost parts of the profile. All textural types of vermiculite ore (massive, stockwork, veins) are well developed. Parent mica, which is phlogopite - Fe-phlogopite, is characterized by a higher Fe content, compared to type 3, f_{com} ranging between 20 and 40, but low F content (lower than 1%). The weathering products of the parent mica are characterized by relatively high f_{com} = 15 - 30. The common exchanged cations are Mg^{2+} - Ca^{2+}.

At the contacts between pegmatite dykes (Belya - Kamak, Vavdos), basic dykes (Gerakini, Zidani) and host ultramafic rocks a zonal alteration is observed. Mica is abundant in a zone around the dykes and sometimes within them. That zone is followed by a vermiculite one. Also, along dislocation contacts vermiculite - rich rock is common.

For the time being the largest vermiculite deposits in the Balkan peninsula (Belya - Kamak, Ichtiman region, Bulgaria) belong to type 4 of vermiculite. The average vermiculite content is 27 %, and average thickness of the weathering crust is 8 meters. The vermiculite reserves are approximately 2.000.000 tons. Vermiculite deposits and occurrences of that type include the Barata and Yarkova Chukara deposits from the above region, the Avren deposits (Rhodope massif) (Bulgaria), Ascos, Vavdos, Gerakini (Serbomacedonian massif) and Zidani in Greece. Commercial deposits of this type of vermiculite have been located in the Russia (Subutakskoe, Kamennie-Mogili and elsewhere) and also, in Texas, USA (Lvova, 1974).

GROUP C, TYPE 5, following the classification proposed fy Lvova (1974), include very small vermiculite occurrences associated with magnesian skarns (Krumovo, Malko Tarnovo area and elsewhwre) and are interest only from the mineralogical point of view.

Hydrobiotite mineralizations in biotite gneisses and schists

(Dolen area, W. Rhodope), can be are classified to **GROUP D, TYPE 9** of vermiculite.

PROPERTIES

The vermiculites from the main type deposits (3 and 4) differ in their properties. The crude vermiculite is mainly contained in the smaller than 3 mm fraction. The loose of weight of the exfoliated vermiculite ranges from 60 to 160 Kg/ m^3 and from 120 to 200 Kg/ m^3 respectively. The two types of vermiculite deposits also differ in their melting point. It is higher than 1350 C for type 3, and 1300 C for type 4. Colour changes during expansion (under the same heating conditions) are dependent upon the type of vermiculite (particularly on f_{com}). After exfoliation the former type turns to silver - white, whereas the latter turn to gold - red - brown colour. Cation exchange capacity of vermiculite ranges from 160 to 210 mgeq/100g and from 120 to 167 mgeq/100g respectively.

AGE OF VERMICULITE DEPOSITS

The determination of the age of the formation of vermiculite deposits is very complicated, due to the long duration of the formation process itself. In an attempt to determine the age of micatization, the K - Ar method was used on parent mica. The analyses were carried out in the Isotopic Geochemistry and Geochronology Laboratoty, in the Moscow University, by E. M. Kolesnikov. The results are given in the Table 2. It is seen that micatization in the Inchtiman region has took place 260 - 300 m.y. ago, e.g. Permian; in the Ascos deposit: 104 - 105 m.y. ago, e.g. Cretaceous . The results for the deposits from the Rhodope massif are quite different, approximatelly 40 m.y. The interpretation of these values concerning the absolute age and its variation is difficult, at the present stage. The tectonic reconstraction and development of the Balkan peninsula has shown several stages of deformation and metamorphism (Karamata et al., 1989). However, the data provided by Lilov et al. (1983); Arnaudov et al (1989) and others, suggest that K-Ar data in the Rhodope massif indicate the time of micatization process, but not the last event of metamorphism.

In summary, ophiolites in the southern part of the Balkan peninsula have been affected by a specific type of weathering (lateritic). The resulting weathering crusts of various morphological types, are rich in Fe, Ni and Co, but they are not vermiculite bearing. Vermiculite - bearing formation are very rare and are related only with a sialitic - geochemical type (kaolinite) of weathering (Lvova, 1974). The geological, structural and petrological data of the Balkan peninsula do not offer evidence to expect the presence of group A of vermiculite. However, there is not doubt that in the Balkan Peninsula are found large deposits with the required features to be vermiculite of acceptable quality. Therefore, it seems likely that the Balkan peninsila offer good possibilities for vermiculite prospecting and exploration.

Table 2. K - Ar isotopic data on phlogopites from vermiculite deposits

Πίνακας 1. Ισοτοπικά δεδομένα K - Ar σε φλογοπίτες από κοιτάσματα βερμικουλίτη

Location	Deposit	Sample	K (wt%)	^{40}Ar (10^{-5})	Absolute age (m.y.)
I	Parapan-	62b/86	7.03	9.46	317 ± 6
h	dovtsi				
t					
i	Barata	588/90	7.56	9.66	302 ± 4
m					
a	Beliya	189sVI	7.18	7.69	257 ± 4
n	kamak	454/88	6.95	7.53	259 ± 3
Serboma-cedonia	Ascos	3/91	8.50	3.56	104 ± 1
Rhodope massif	Seltse	2/90	7.73	1.20	40 ± 1
	Kostandovo	2/89	5.72	0.70	32 ± 1

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