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MINERALOGICAL–GEOCHEMICAL STUDY OF URANIUM BEARING GRANITE PHASES IN PARANESTI AREA, N. GREECE

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Abstract: This study concerns the petrological-geochemical characters of the “granite type” rocks from Paranesti area, in which I.G.M.E. has localized the most important uranium ores in Greece. Their mineralogical phases are examined and they are correlated with the geochemical data of the major elements, as well as with some of the trace elements from mineralized samples of the area.

Key words: Mineralization, Dipotama, Fteroto, Spilia

1. Introduction

In this paper the petrological characters and the uranium content of the Meta-, or Middle- Hercynian age, (according to different authors), “granite type” rocks from the inner parts of the Rila–Rhodope granite area are examined, and especially from the small granite body which is located in the southern part of this granite mass and it is known as “Paranesti granite”. More specific, the aim of the study is a first effort to define correlations between petrographical–geochemical characters of the three granite phases and the uranium mineralization types, so that it will be possible to define the whole metallogenetic circle of uranium in the Paranesti area (Drama, N. Greece).

On the basis of petrographical criteria, in combination with the classification of the uranium deposits, (Pergamalis et. al., 1998), classified these rocks into three different phases: a) the granite phase of Fteroto (ores of Fteroto and Spilia), b) Prasinada phase and c) Dipotama phase (ore of Archodovouni), which respectively occupy the upper, the middle and the lower granite part. As the granite phases, the uranium ores also are distinguished into three types, the reduced, the oxidized and the mixed or “complex” type ore.

2. Geology

The area of Paranesti belongs to the Rhodope complex. It consists of a) late Cambrian age metamorphic rocks, b) Meta-, or Middle- Hercynian age granite bodies (which are known as granite of Skalotis and granite of Paranesti), included inside them and c) an Oligocene age volcanic cover of rhyolites, quartz diorites, aggregated tuffs and tuffites gradually changing in sediments of the underlying volcanosedimentery series.

The uranium ores are located in the north part of the “Paranesti granite”, although disperse metallogenetic indications (which do not participate in the main uranium ores) have been localized inside the volcanic cover and the volcanosedimentery series.

The geological map of this area (in a 1:5.000 scale) was checked geometrically on the surface, as well as in depth, by F. Pergamalis et. al. (2001) and the existence of three different granite phases in an harmonic bedding correlation was established, (Fig. 1a,b):

- The upper granite phase, or the “Fteroto type”.
- The middle phase, or the “Prasinada type” and
- The lower granite phase, or the “Dipotama type”.

The uranium ores, (which have been studied till today) are also distinguished in three categories (Pergamalis et al., 1998):

- The endogranitic type containing the reduced pitchblende uranium ore. The body 1 of Spilia, Paranesti is a representative example of this category.
- The endogranitic type containing the oxidized uranium ore with secondary U^{6+} phosphoric salts, especially spherolithic Renardite. The Fteroto ore body is the most representative example of this category.

- The mixted (or complex) endogranitic type (containing oxidized and reduced uranium ore), with U^4 and U^{6+} phosphoric and silicate salts (coffinite, autunite), creating filons or amass of ore. The Archodovouni ore is the most representative example.

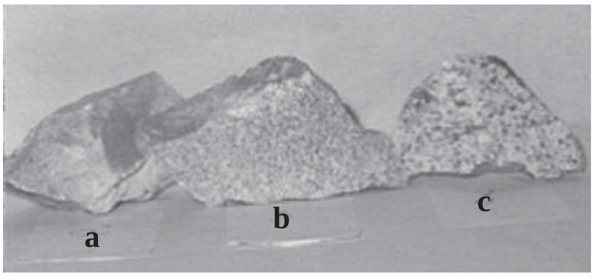

Fig. 1a. The three different "granite types" of Paranesti area rocks.

The first two categories of uranium ores were located in the upper granite phase, the "Fteroto type" granite, the third category was located in the lower granite phase, "Dipotama type". Till today inside the middle granite phase, the "Prasinada type " any uranium ore have not been located.

3. Mineralogical Characters

This mineralogical study, as well as the following geochemical study, refers to samples from the two uranium bearing phases of Paranesti granite, a) the upper phase or "Fteroto type granite" with two representative ore bearing bodies, (the Spilia and the Fteroto ore bodies, photos 1-4 and 10-12), and b) the lower granite phase or "Dipotama type" with the Archodovouni ore as representative example (photos 5-9).

3.1. Upper Granite Phase, or "Fteroto Type"

The most important and richer uranium ores are found in this granite phase, which is a microgranular, leucocratic rock, consisting mainly of quartz and feldspars.

3.1.1. Granite from Spilia ore body

The hosting rock is high tectonized, with cataclas-

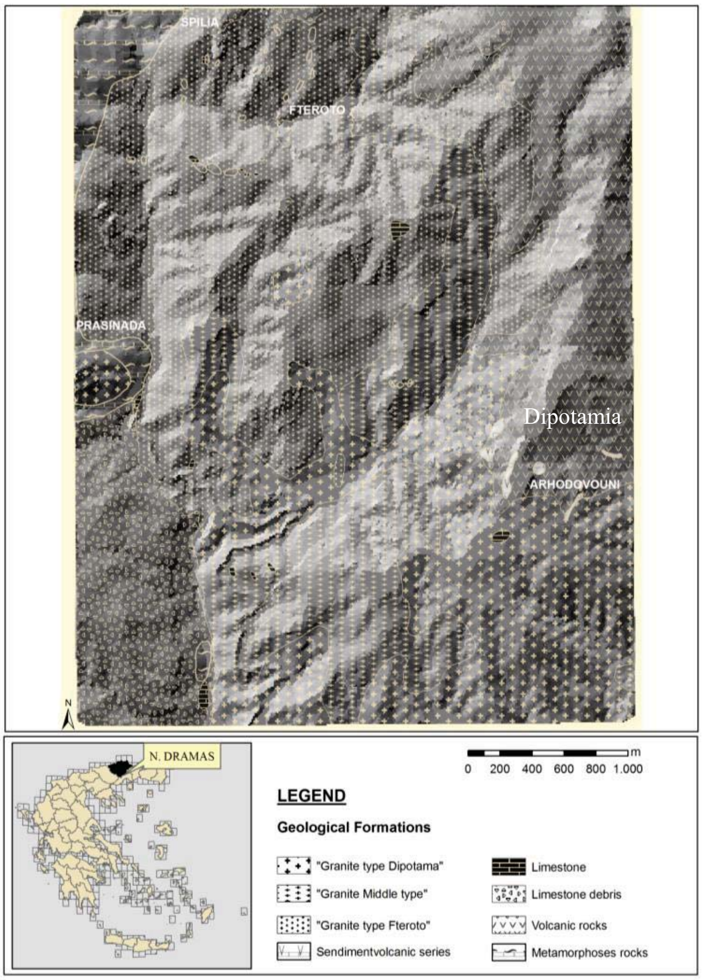

Fig. 1b. Paranesti's geological mapping.

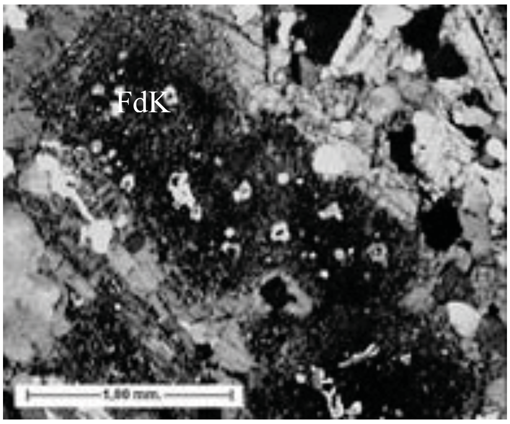

Photo 1. // Nickols, “Fteroto Granite type” from Spilia Paranesti ore. Completely altered alkali feldspar, with only a fine rim of unaltered crystal, (albitization, calcitisation, sericisation etc.) remaining.

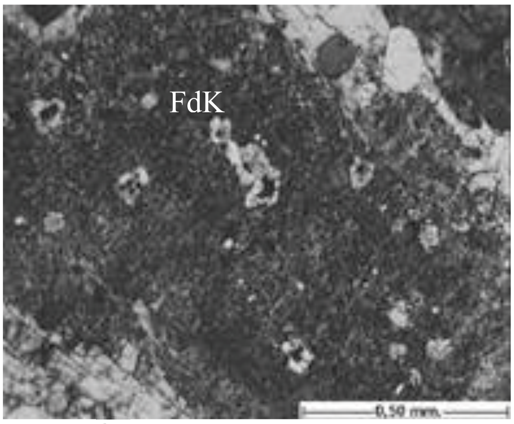

Photo 2. ⊥ Nickols: Details of the photo 1.

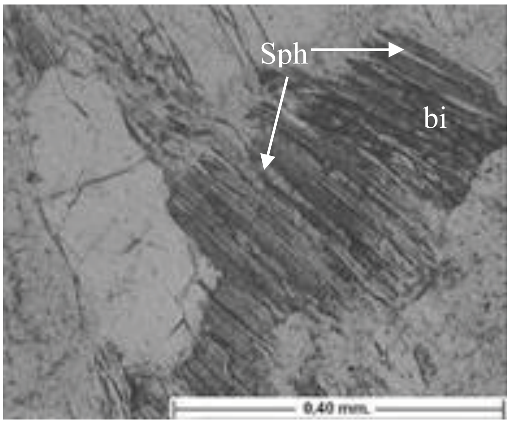

Photo 3. // Nickols: “Fteroto Granite type” from Fteroto ore Paranesti. Green biotite, with inclusions of sphene along the cleavage surfaces.

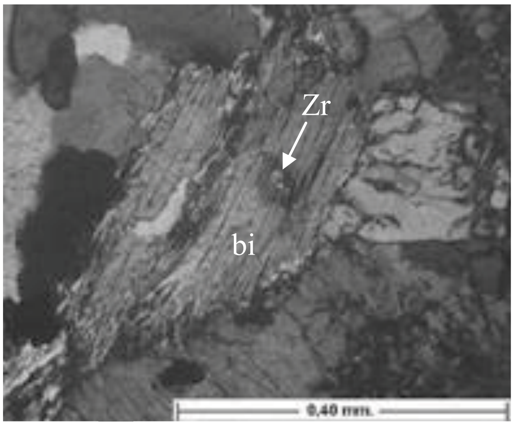

Photo 4. ⊥ Nickols: Fteroto “Granite type”, Fteroto ore. Inclusions of zircon crystals, with pseudochroic halo, in an unstable biotite presenting a zone of neoformed crystalline minerals.

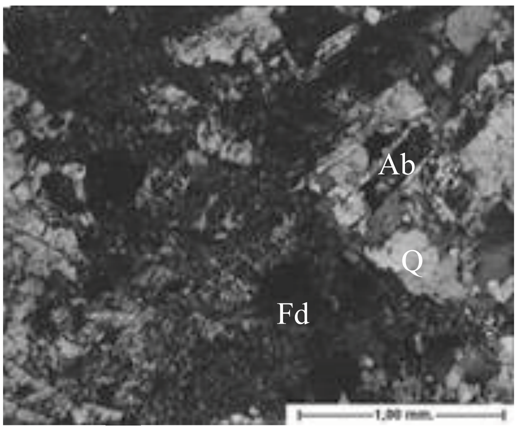

Photo 5. ⊥ Nickols: “Dipotama Granite type”, Archontovouni ore, Paranesti. Feldspar crystal with tectonic strain, and a neoformed fine crystalline material, consisting of quartz, albite and sericite.

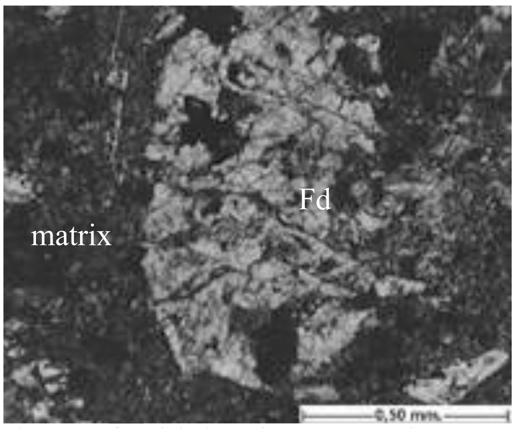

Photo 6. ⊥ Nickols. Dipotama Granite type”, Archontovouni ore, Paranesti. Islands of tectonically strained feldspar crystals, dispersed inside fine granular, neoformed quartzofeldspathic material.

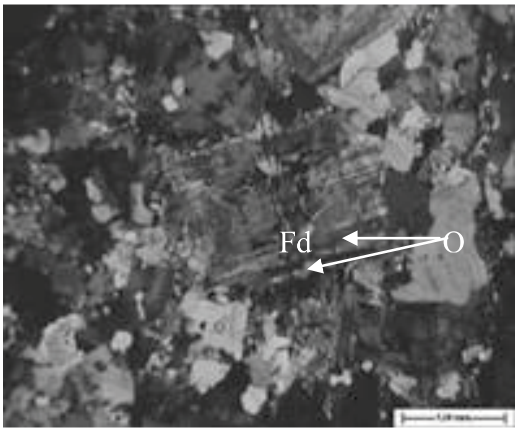

Photo 7. //Nickols. "Dipotama Granite type", Archontovouni ore, Paranesti. Feldspar crystals with altered zones along the tectonic fractures, surrounded by small recrystallized quartz grains.

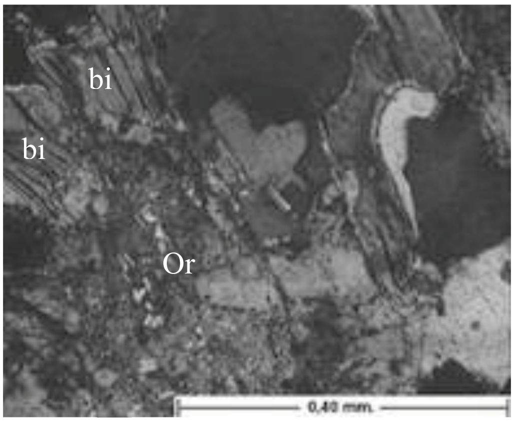

Photo 8. ⊥ Nickols. "Fteroto Granite type", Fteroto ore, Paranesti. Strongly altered orthoclase (sericitized, alterations in clay minerals) and corroded biotite leaflets with tectonically slightly deformed, (curved), cleavage surfaces.

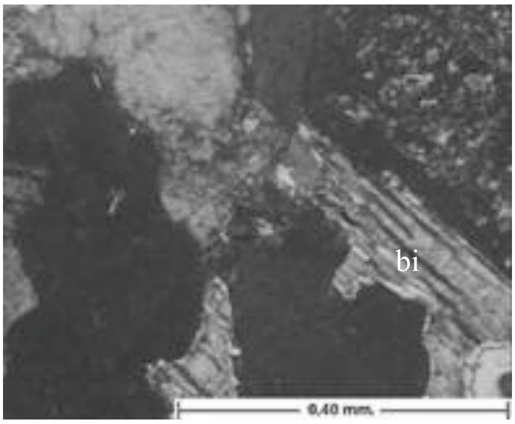

Photo 9. Detail of Photo No-8.

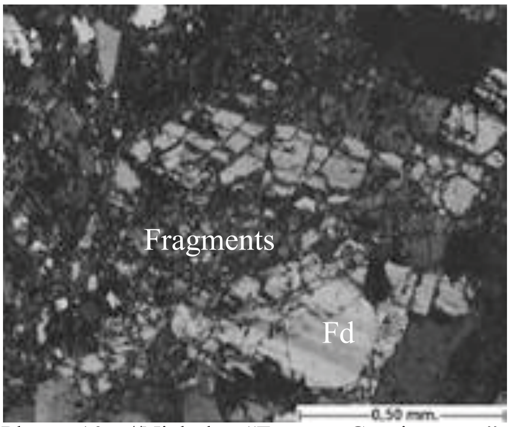

Photo 10. //Nickols. "Fteroto Granite type", Spilia ore, Paranesti. Completely fragmented feldspar crystal, with fine quartzofeldspathic material between the fragments.

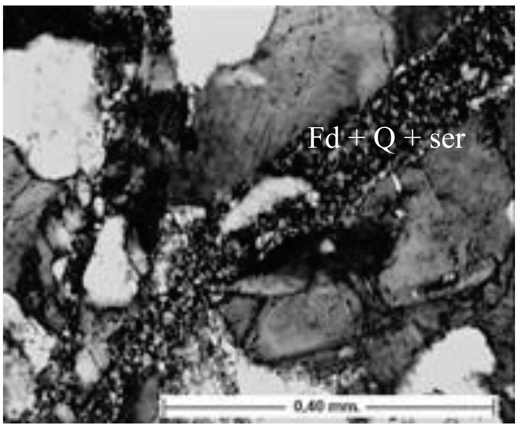

Photo 11. //Nickols. "Fteroto Granite type", Spilia ore, Paranesti. Zone with a fine crystalline mixture of quartz, feldspars and just a little sericite, cutted by another one zone with more diffuse boundaries, consisting of small recrystallized, granular quartz.

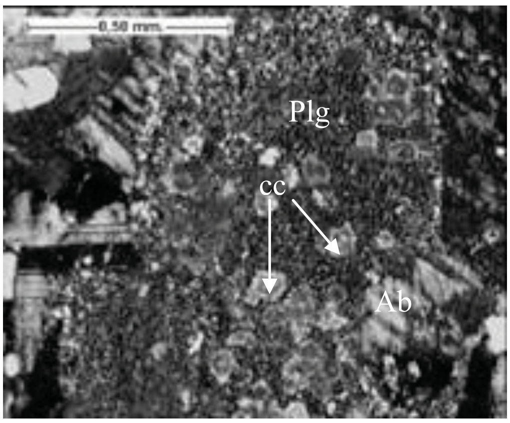

Photo 12. ⊥ Nickols. Fteroto "Granite type", Spilia ore, Paranesti. Detail. Strongly altered plagioclase, with albitization and calcitization forming disperse nests of calcite and albite.

tic texture, sometimes presenting a dense, thick network of intercrossed veinlets, crossing the rock at different directions and sometimes forming parallel or intercrossed zones of fine granular quartzofeldspathic material. Inside this matrix fragments of quartz and feldspars crystals are found, that constitutes up to 40% in volume of the rock. Between the zones the rock presents cataclastic texture and it is consisting of quartz, alkali feldspar and plagioclases, all about at the same proportion.

The quartz forms allotriomorphic crystals, the alkali feldspars present alterations into calcite, sericite and clay minerals, while the plagioclases present extended albitization, whith only a peripheral skeletal halo of plagioclase remaining, the neoformed idiomorphic albite occupies up to 80% of the crystal volume,. In some plagioclases the neoformed albite coexist with calcite concentrations, either inside sparse small nests, either inside a small irregular network of filons.

Except these major minerals that constitutes 90 to 97% of the rock volume, are also found uncolorized biotite leaflets presenting deformed (curved) cleavage surfaces, that usually include zircon and apatite, and present a small degree chloritization. As accompany minerals zircon, apatite, idiomorphic crystals of an opaque mineral are found, and also, inside the zones with the cataclastic texture, concentrations of an amorphous opaque mineral.

3.1.2. Granite from Fteroto ore body

The hosting rock, of the Fteroto ore body, is a tectonized and partially recrystallized leucogranite. Quartz, alkali feldspars and plagioclases (the three about at the same proportion) constitute about 95-97% of v. of the rock. Biotite and white micas are also presents in the same proportion, as well as, zircon and small grains of a black opaque mineral, as accompany minerals.

The plagioclases and the alkali feldspars are, at lesser or bigger degree, altered in albite, sericite and clay minerals. The secondary calcite concentrations inside the irregular veinlets with dentoidal rims show that, during the secondary process of the rock, was taken place Ca dissolution from the plagioclases. The quartz crystals are usually broken and often recrystallized, forming quartz nests between the bigger quartz crystals and the feldspars, while the biotite is usually uncolorized and chloritized.

3.2. Lower Granite Phase, or "Dipotama Type"

It is a coarse grained granite, consisting of quartz, alkali feldspars (orthoclase and microcline), plagioclases (oligoclase), mica, (biotite and white mica). The first three minerals are found almost in the same proportion and constitute the 95% of v. of the rock forming allotriomorphous crystals, which can be considered as belonging in two different generations, the first with bigger crystals and the second with much smaller ones.

The quartz is found either in big xenomorphic multi-crystal aggregates, or in smaller grains covering spaces between the feldspars. The plagioclases and the alkali feldspars appear altered at a smaller or bigger degree, forming albite, sericite, clay minerals and calcite, often all coexisting in the same crystal.

The rare and small biotite leaflets are uncolorized and they present opaque inclusions of titanium and of semitransparent iron oxides. As accompany minerals zircon and apatite are present. Also rarely inside this rock, a few, thin zones, consisting of very fine, granular, quartzofeldspathic material and small fragments of albite, appear.

4. Geochemical Characters

The the Paranesti granite type rocks present a strong potassium–sodium character, due to the abundance of feldspars (alkali feldspars and plagioclases), while the biotite participates only in a very small percentage. According to the terminology of Bayly (1976), these rocks could be characterized as granite with biotite, or oligobiotite quartz porphyritic granofelsite, so, it will be possible to overpass some indications about the origin provided by the term granite.

According to the same author, to define the magmatic origin for magmatic or metamorphic rocks, when the discrimination is too difficult or impossible, the only visible characteristic should be the great homogeneity of the rock on a vast surface, bigger than 5 Km^2.

60 samples, representing all petrological types in the studied area, were analyzed by the X.R.F method. (X-ray fluorescence spectrometry), for both, the main and trace elements, with a Philips automatic sequential spectrometer PW 1450.

4.1. Main Elements

To understand the chemical changes of the main elements and for the classification of the different samples (Figs. 2, 3, 4), the results of the chemical analysis were projected on different diagrams.

The changes of the $K_2O/N\alpha_2O$ ratio value are significative (Figure 2) and they are mainly the result of the $N\alpha_2O$ changes, because the K_2O content remain constant, except for “Dipotama granite type”, which presents a simultaneous increase of K_2O-$N\alpha_2O$ that characterises the uranium rich granite types (from 84 to 3.740 ppm of U).

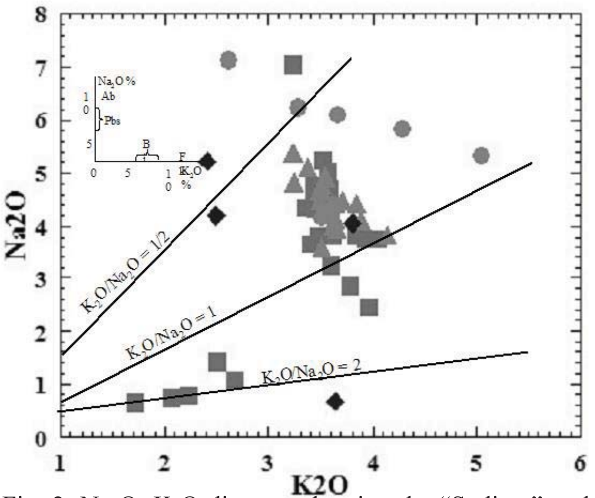

Fig. 2. Na_2O, K_2O diagram showing the “Sodium” and “Potasium” character of the Paranesti “granite type” rocks. The lines ½ and 2 limit these characters, while the increase of the uranium content follow different tendencies for each area. In the small diagram are projected the compositions of the main sodium and potassium mineralogical phases.

LEGEND for all Figures

■ Archodovouni ore ▲ Spilia 2 ore

⬣ Fteroto ore ◆ Spilia 1 ore

The fluctuations of the $K_2O/N\alpha_2O$ ratio are significant when varying in the interval from 4,0 to 0,3 and, when the biotite is present in a small or minimum percentage, in some way, this fluctuation reflects the rapport between potassium feldspars / plagioclases, or, in the case of a major $N\alpha_2O$ change versus the K_2O content, the preponderance of sodium terms in the potassium–sodium mineralogical phases (formation of albite), which in the case of Archodovouni, and in a smaller degree, in the case of Spilia area, are correlated with the enrichment in uranium.

The “Dipotama type” granite, (ore of Archodovouni), presents the higher values in $K_2O/N\alpha_2O$ ratio, (from 4 to 1). The “Fteroto type”, (ore of Fteroto), presents the lower values, (from 1 to 0,3), while the “Fteroto type”, (ore of Spilia), presents intermediate values, with members from the higher to the lower value. In each granitic type, (sometimes even inside the same type), the uranium enrichment follows different tendencies. For example, in contrast with the “Dipotama type”, the “Fteroto type” presents a uranium enrichment (from 54 to 787 ppm) with a great diminution of the sodium content (Na_2O) compared that of the potassium (K_2O) content, (a mean value of 1,67 against 0,78, respectively). The Spilia ore granitic type presents also a double evolution, firstly the decrease of the $K_2O/N\alpha_2O$ value, either with a rise or either a decrement of the uranium content at the different samples (Fig. 3).

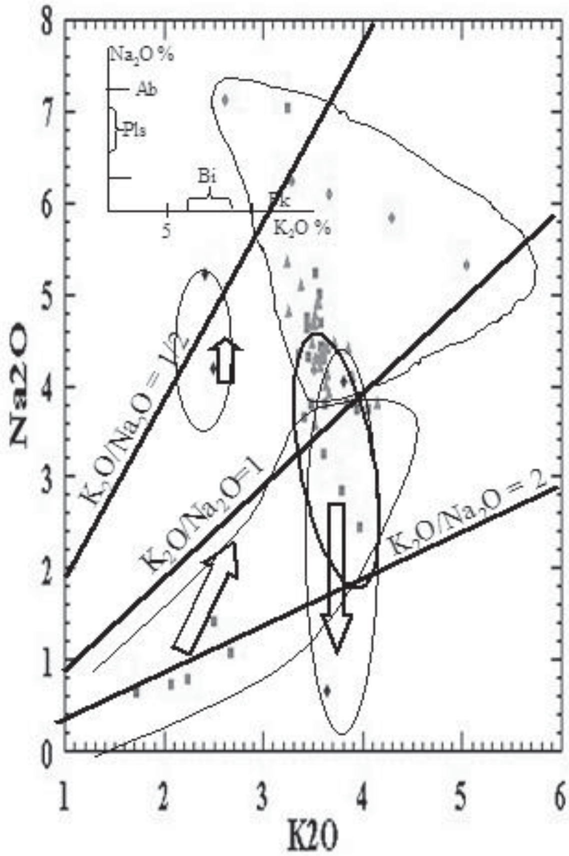

Fig. 3. Na_2O, K_2O diagram showing the variations of the Na and K content in correlation with the increase of the uranium content in the Paranesti ore bodies.

In the K_2O–SiO_2 diagram of Peccerillo and Taylor (1976), the granite type rocks of Paranesti with mineralization, are placed in the areas of the potassium rich andesites–dacites (high K), of the potassium calc-alkaline series, with the acid members occupying the areas of dacites–rhyolites, at the borders between the potassium calc-alkaline series with the calc-alkaline one (Fig.4).

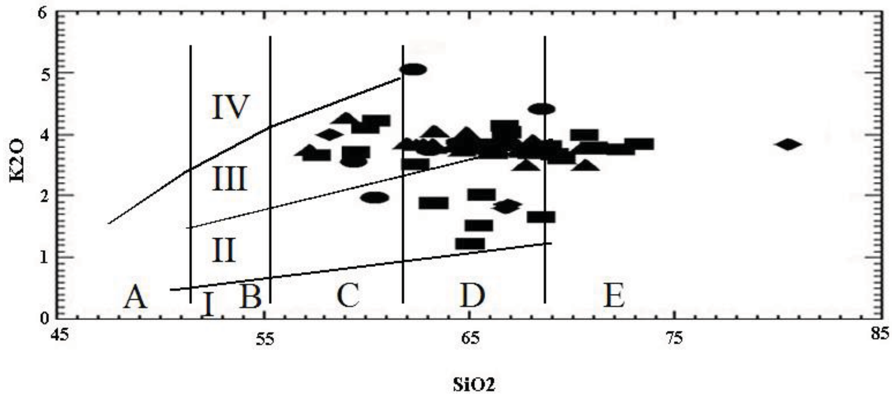

Fig. 4. Projection of the "Granite type" rocks from the Paranesti area ore bodies at the diagram of Peccerillo et Taylor (1976). The borders of the different series correspond to the axis of K_2O and these of the relative volcanic rocks to the axis of SiO_2.
I. Tholeitic series.
II. Calc-alkaline series, (D: dacite, E: rhyolite).
III. Potassium Calc–Alkaline series, (C: Rich in K andesite, D: Rich in K dacite).
IV. Soshonitic series.

The "Dipotama granite type" (ore of Archodovouni), shows the bigger dispersion, with mineral bearing samples in which the potassium content is slightly reduced from the alkali to the acid members, in contrary with the evolution of the unaltered rocks. The other granite types of Fteroto and Spilia ores, present also the same evolution, but with a slightly lesser fluctuation.

The volcanic rocks of Paranesti (biotite–hornblende rhyolites and the more alkaline augite–hornblende quartz diorites, Soldatos, 1961), are also interesting. These also are always strongly enriched in potassium and they are placed at the acid end of the potassium calc–alkaline series, but without the less acid derivative members. Especially, the accumulation of Paranesti area volcanic rocks, towards the borders of the acid K-calc-alkaline and the acid soshonitic series, gives the impression that the volcanic rocks of Paranesti must be the last derivative members (rich in SiO_2 and K_2O) of volcanic materials strongly enriched in potassium, namely members of a high-potassium calc–alkaline to soshonitic series, which is impossible to be determined exactly without the analysis of the more basic members. Generally the volcanic rocks of Paranesti area present chemical characters of a high-potassium continental border environment (Jakes and White, 1972). The chemism of these rocks does not differ essentially from the granite type rocks originated from the same zone.

At the diagram Na_2O+K_2O/SiO_2, (Fig. 5) all the mineralized granite type samples of Paranesti are projected in the area of the Kuno's alkaline series, (Richard, 1988-1995). The alkaline character presents a decreasing tendency from the Fteroto area samples to the samples of Spilia and Archodovouni area. This evolution, at the beginning, is slightly positive or constant with a limited drift, and then, for the mineralized samples of these Paranesti areas, becomes negative. Some samples from Spilia and Archodovouni areas tend to be integrated at the extension of the calc-alkaline series of Kuno.

Except all these characters, which are directly related with the changes between potassium and sodium terms of the feldspars, (albitization), the TiO_2 content of Paranesti area granite type rocks, is low, (from 0,03 to 0,29%) and only some samples present contents from 0,41-0,56%. The total iron $(FeO)_{Tot}$ present a bigger variation from 0,10 to 5,10%, with the uranium richer granite types (U content from 500 to 4.693 ppm), associated with an $(FeO)_{Tot}$ content greater than 1,20%.

The same correlation presents also the magnesium for contents bigger than the 0,17% in MgO, but keeping however a lower fluctuation from 0,10 to 1,96 %.

The manganese and the phosphor fluctuate in very low levels, from 0,01 to 0,40% for MnO and from 0,02 to 0,19% for P_2O_5 without any association with the uranium content. Finally, the calcium presents low variations, from 0,32 t0 1,89% in CaO, except for some samples of Spilia area, presenting contents from 2,65 to 4,38% in CaO and characterizing uranium-rich granite type (2.763 to 3.731 ppm in U).

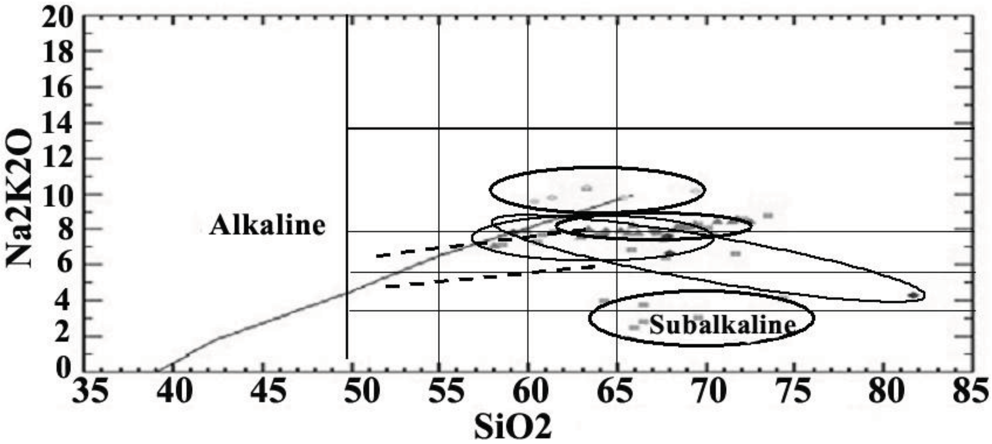

Fig. 5. Diagram SiO_2-(Na_2O+K_2O) showing the projection of the mineralized samples from Paranesti area at the diagrams of Kuno (1960) and Cox et al.(1979), (L. R. Richard, 1988-1995).

4.2. Trace Elements

After the main elements, we will also study the trace elements to see if they certify or they complete the interpretations related with the typology, as well as, with the evolution of the Paranesti-Drama area granite type rocks.

Twelve (12) trace elements (Ba-Rb-Sr, Zr-Y-Nb, U-Th και Co-Ni-Cr-V) were analysed by X Ray Fluorescence (XRF) method in the laboratories of Mineralogy-Petrology Department of I.G.M.E.. From all these elements, only the concentrations of Ni were lower than the instrument detection limit.

The Ba-Rb-Sr diagrams will accomplish the trace elements study for the certification of the geochemical, petrographical and typological correlations. In the ternary diagram (Fig.6) of El Bouseily et al. (1975), five different zones (the diorite, the granodiorite, the quartz-diorite, and the anornal, normal and very differentiated granites) can be seen. Most of the Paranesti granite type rocks are projected at the granodiorite-quartz diorite area. Some fluctuations between Ba-Sr and also between Ba-Sr-Rb reflect the ratio between alkali feldspars/ plagioclases and the prevalence or not of the first component over the second (case of albitization).

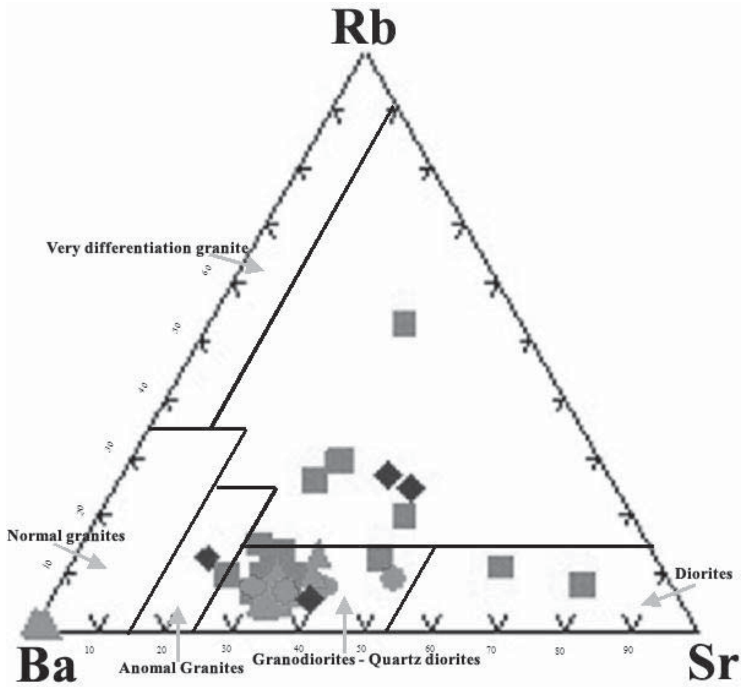

Fig. 6. Projection of the "granite type" rocks from Paranesti-Dramas' area at the diagram Sr-Rb-Ba.

The Paranesti-Drama area, granite type rocks, in the diagram Rb/Sr (Fig.7) of Johan et al. (1980), which defines different magmatic series, (orogenetic zones, endocrystaline granitic mass and andeitic shossonitic series) are also projected in the area of the soshonitic – potassium calc-alkaline series.

The uranium content of the Paranesti area granite type rocks, present a wide range of values, from 19 to 4.693 ppm, and, according the mineralogical criteria concerning the uranium, they belong to the category of ore samples. In particular, from the total number of samples, only 6 present a content lower than 8 ppm, (the double of the middle granite content), 14 samples a content from 8 to 100 ppm, 27 samples from 100-500 ppm and 12 samples have a content higher than 500 ppm.

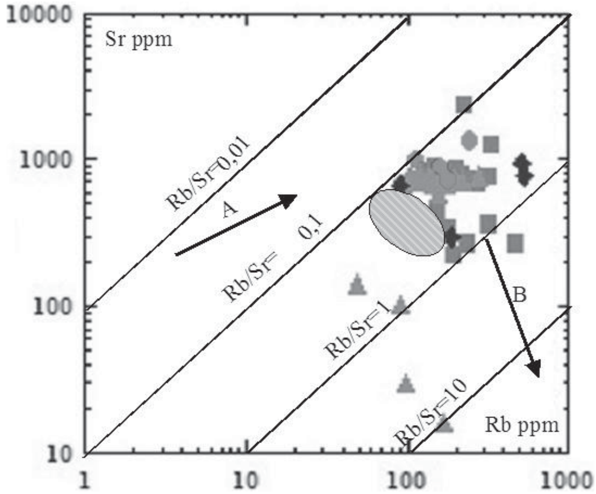

Fig. 7. Projection of the "granite type" rocks from Paranesti, Dramas' area, at the Rb-Sr diagram limiting the different magmatic series The tendencies A and B show the characteristics orientation of the orogenetic zones and the endocrystalline granitic masses respectively. The shaded ellipsoidal area shows the champ of the andesitic and soshonitic series (Johan et al. 1980).

Respectively, the content in thorium varies slightly, with values between 4 to 78 ppm, so, the ratio Th/U is very low, from 0,001 (mainly for samples from Archodovouni area), till 1,0 (mainly for samples from Spilia area), while, intermediate values of Th/U ratio, (between 0,01 and 0,1), presents the Fteroto area (Fig.8). For some samples of the Fteroto area and for a U content around 200 ppm, appears a slight correlation of the uranium increase with a smaller increase in Th, but for most samples the increase in U is accomplished with a constant Th content.

If we consider the quotient K/Rb (Fig.9), as a sign of the rocks evolution, we observe a considerable increase of the uranium content, from 10 to 1.000 ppm, in a strong decrease of that quotient (from 300 to 30).

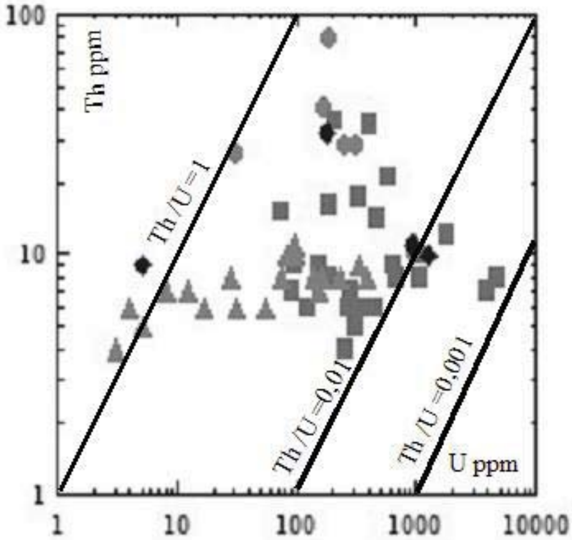

Fig. 8. Correlation of the Uranium and Thorium contents of the "granite type" rocks from Paranesti, Dramas' area.

The samples from Archodovouni area and some from Spilia area, present the lower K/Rb quotient, with the higher U contents, the same time that the higher K/Rb quotient is present in some samples from Fteroto (ore Spilia), granite type rocks, with the smaller uranium content. An intermediate place occupies the Fteroto ore, with middle values of K/Rb quotient and U content.

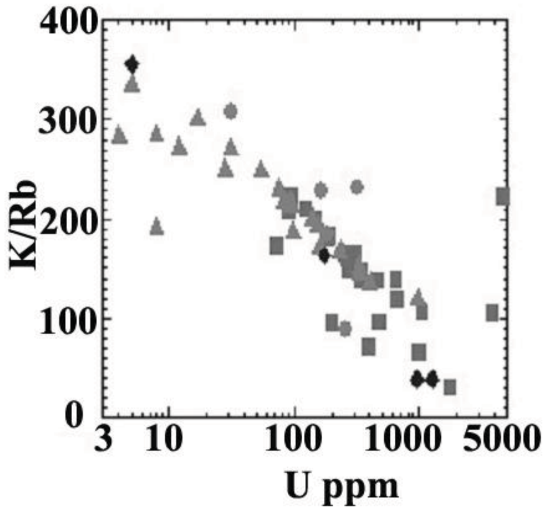

Fig. 9. Correlation between the uranium content and the K/Rb quotient, in the different ore bodies from the Paranesti area, upper and lower "granite type" rocks.

The contents in Zr-Nb-Y, as well as these in Co-Ni-V-Cr, fluctuate in low levels, without any correlation with the increase of uranium content, while the higher contents of the second group trace elements, are present for the lower contents in uranium (<8 ppm in U).

5. Conclusions

The middle- or meta-hercynian age, granite type rocks of Paranesti area, are known for the most important uranium deposits, which have been localized in Greece until today. Therefore in this study, has been performed a first effort to define the nature of the rocks hosting the U mineralization, and to define correlations with the petrographical–geochemical similarities and differences that they present between them and with their uranium content.

The geochemical characters of the mineralized rocks from Paranesti area, are the similar with these of the potassium – calc-alkaline series, with highly enriched in potassium terms, even for the less acid members. Like the volcanic rocks of the cover layer, they present characters of a high potassium continental margin environment.

The two U mineralized granite types of Paranesti area, (which have been geologically charted in previous studies), present characteristic fluctuations of the K_2O – Na_2O ratio, related with the changes of the Na_2O content and indicating the formation of the sodium terms (albite), in the potassium–sodium mineral phases (alkali feldspars - plagioclases). In the case of Archodovouni area, but also at a smaller degree in the case of Spilia area, this formation is connected with the enrichment in uranium, preserving constant the thorium contents. This last element shows an lack comparing to the U, so the ratio Th/U is very low, from 0,001 to 1, in contrary with the contents of the above elements in the granites (Th/U≈4). The "Fteroto type" granite is just a little different, with a small correlation in the uranium content increase, with a clear smaller increase of the Th content.

The "Dipotama type" granite, (ore Archodovouni), with the less accentuated alkaline character, presents the higher values of K_2O/Na_2O ratio, with high K_2O contents, even for the less acid terms, and with higher U contents for the most differentiated types (of the lower K/Rb quotient).

The "Fteroto type" granite, (Spilia ore), with an alkaline character similar to that of the Archodovouni ore, presents intermediate values of K_2O/Na_2O ratio, with a high K_2O content, even for the less acid terms and some of its samples, from the most differentiated types, have the highest uranium contents, and some other, less differentiated samples, (with the higher K/Rb quotient), the smaller contents in U.

Intermediate K_2O/Na_2O quotient and uranium contents present the Fteroto ore, (granite "type of Fteroto"), with the more accentuated alkaline character.

From the rest of the elements, Fe and Ca (oxidization–calcitization) show a slight relation with the uranium enrichment, while the trace elements Ba-Rb-Sr certify and complete the conclusions of the main elements.

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