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GEOHEMIJSKE KARAKTERSISTIKE UZORAKA SA DOMINANTNOM ALUNITSKOM FAZOM IZ LECKO-RADANSKOG VULKANSKOG KOMPLEKSA (SRBIJA)

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Ključne reči: aluniti, mikroelementi, elementi retkih zemalja, Lecko - Radanski vulkanski kompleks

U ovom radu prezentovan je sadržaj mikroelemenata (As, Ba, Co, Cr, Cu, Ga, Ge, P, Li, Nb, Zn, Ni, Pb,V, Zr i Sr) i elemenata retkih zemalja (REE - lantanidi, Y, Sc, Th) u šest odabranih uzoraka sa dominantnim učešćem alunitske faze (>50%) i utvrđene su osnovne korelacije između njih. Sadržaj mikroelemenata i elemenata retkih zemalja (REE) određen je ICP/OES tehnikom (Spectro Blue, Germany), posle totalnog razlaganja topljenjem sa $NaKCO_3$ na 1000 °C. Ovo istraživanje predstavlja nastavak istraživanja pojava alunita u Lecko-Radanskom području. Tokom prethodnog perioda istraživanja (2009-2013) definisan je mineraloški i hemijski sastav ovih uzoraka (Tančić et al., 2021), kao i procentualno učešće minerala alunitske faze, odnosno alunita $KAl_3(SO_4)_2(OH)_6$, (27-52%), natroalunita $NaAl_3(SO_4)_2(OH)_6$, (27-48 %) i šlosmaherita $HAl_3(SO_4)_2(OH)_6$, (5-46 %) . Pored minerala alunitske faze, u ovim uzorcima detektovano je prisustvo dominantnog kvarca (u 5 uzoraka), a takođe i manje zastupljenih kaolinita (samo u 2 uzorka) i tridimita (samo u 1 uzorku).

U poređenju srednjih i pojedinačnih vrednosti, sa vrednostima sadržaja datim za kontinentalnu koru, može se zaključiti da su srednji i pojedinačni sadržaji As, Pb, Ba, Sr i Li, povišeni u svih šest uzoraka. Srednji sadržaj La je povišen u odnosu na referentnu vrednost u svim uzorcima osim u jednom uzorku. U istom uzorku je sadržaj većine mikroelemenata minimalan (izuzev povišenog sadržaja Ga), najverovatnije zbog toga što je alunit formiran u zoni sa najvišom temperaturom. Sadržaji Ga i V, koji premašuju referentne vrednosti u tri uzorka, najverovatnije ukazuju na obogaćivanje alunita ovim elemenatima usled prolaska hidrotermalnog fluida kroz ugljonosne sekvence, pri njihovom formiranju. Na istražnom području ovo prisustvo se odnosi na sediment donjeg i srednjeg miocena, bogatog organskom materijom (Čučalska sekvenca).

Pored navedenih elemenata, uzorci sa dominantnim učešćem alunita su generalno obogaćeni elementima La, Ce, Nd (laki elementi retkih zemalja – LREE) kao i Th i Sc. Visoke vrednosti koeficijenata značajne korelacije ($r > 0,60$) zbira sadržaja LREE - ∑LREE sa Al ($r = 0,91$) i sa P ($r = 0,65$) mogu ukazati na to da su LREE, posle hidrotermalnih alteracija ušli u strukturu alunita, pre nego HREE (teški elementi retke zemlje). Dodatno, značajna korelacija ∑HREE sa Zr ($r = 0,70$) i Sc ($r = 0,69$), može se objasniti njihovom manjom mobilnošću i većim afinitetom vezivanja za refraktorne minerale, otpornije na dalje alteracije (Ramos et al., 2016). Vrednosti koeficijenta značajne korelacije Nb-Th ($r = 0,61$), Nb-P ($r = 0,91$), Nb-Al ($r = 0,60$), Nb-∑REE ($r=0,61$), ukazuju na njegovo prirodno grupisanje sa ovim elementima (Hawks and Webb, 1968) i u slučaju alunita. Vrednosti sadržaja Li proporcionalno rastu sa porastom K u odnosu na prisustvo Na/H alunita (natroalunit/šlosmaherit), $r=0,85$ u uzorcima, što sugeriše da je Li, prilikom formiranja mogao menjati najpre K u kristalnoj strukturi alunita.

GEOCHEMICAL CHARACTERISTICS OF SAMPLES WITH DOMINANT PRESENCE OF ALUNITES FROM THE LECE-RADAN VOLCANIC COMPLEX (SERBIA)

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Key words: alunites, trace elements, rare earth elements (REE), Lece-Radan volcanic complex

The study represents the composition of minor elements, including rare earth elements (REE) in six selected samples with a dominant alunite content (>50%), having a significant correlation between them. The content of the trace (As, Ba, Co, Cr, Cu, Ga, Ge, P, Li, Nb, Zn, Ni, Pb, V, Zr i Sr), and REE (lanthanides, Y, Sc and Th) elements, is characterized using by ICP / OES technique (Spectro Blue, Germany), after total fusion with $NaKCO_3$ at 1000 °C. During the period 2009-2013., a geological study of alunite occurrence in the Lece-Radan area was performed. The mineralogical and chemical composition of major components of the samples is characterized in this phase (Tančić et al., 2021), as well as the composition of minerals within alunites group: Alunite $(KAl_3(SO_4)_2(OH)_6$, (27-52%), Natroalunite $NaAl_3(SO_4)_2(OH)_6$,(27-48 %), Schlossmacherite $HAl_3(SO_4)_2(OH)_6$ (5-46 %). Besides alunite mineral group, in these samples are detected dominant presence of Quartz (in five samples) , Caolinite (two samples) and Tridimite (one sample).

By comparison of the mean and the single values of the trace elements with the reference - the continental core contents, we conclude that content of As, Pb, Ba, Sr and Li are with elevated values. The average content of La registered across of all samples used for this study, with exception of the one sample. This sample is characterized with the lowermost content of the most trace elements explored (with exception of the content of Ga). Most likely, such situation is a consequence of the formation of the alunite in this sample that is collected within the zone the highest temperature (see Tančić et al., 2021). The content of Ga and V, which exceed the reference values in other two samples as well, could indicate the enrichment of the alunite with these elements. This could be a result of the interaction of a hydrothermal fluid, formerly moving along or near a coal-bearing stratigraphic sequence. Indeed, the study area contains a number of organic-type carriers, including a documented presence of the Lower and Middle Miocene sediments, rich in organic matter (Čučalska sequence). In addition, the analyzed samples are enriched with the elements La, Ce, Nd (group of Light Rare Earth Elements - LREE), as well as Th and Sc. The significant correlation coefficient ($r > 0.60$) of the sum of LREE -∑LREE contents with Al ($r = 0.91$) and with P ($r = 0.65$) may indicate that LREE, after hydrothermal alterations, have entered into the alunite structure, characterized with a higher affinity than HREE (Heavy Rare Earth Elements). Additionally, higher value of the correlation coefficient ∑HREE with Zr ($r = 0.70$) and Sc ($r = 0.69$), can be explained by their lower mobility and higher affinity for refractory minerals, that are more resistant to further alterations (Ramos et al. 2016). A significant correlation between Nb-Th ($r = 0.61$), Nb-P ($r = 0.91$), Nb-Al ($r = 0.60$), Nb-∑REE ($r = 0.61$), indicate its natural grouping with these elements that are present (Hawks and Webb, 1968) in the alunite structure itself. The content of Li is in the function of the alunite presence (%), associated with the Na- and H- alunite (Natroalunite and Schlossmacherite, respectively ($r = 0.85$). Such correlativity indicates that during the mineral formation, K was rather exchanged by Li in the crystal structure of alunite.