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DISTRIBUTION OF RARE EARTH ELEMENTS IN METAMORPHIC ROCKS OF PLATTENKALK GROUP AT CENTRAL CRETE, GREECE

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Abstract

Within the metamorphic rocks of the Plattenkalk Group, in an occurrence at Tallaia Mountains, central Crete, light rare earth elements Cerium and Lanthanum are found. The studied rare earths are seen in an epidote group mineral, allanite. Scanning Electron Microscopy of the samples showed clearly the crystals of allanite (pre-kinematic crystals) and their grain size. Mineral processing was conducted for the size fractions: (<10 μm), (10-212 μm) and (212-1000 μm), according to the magnetic properties and grain size of allanite. After magnetic separation of the fraction (<10 μm) and (212-1000 μm) rare earths Ce and La were observed to have higher concentrations in the magnetic products. In conclusion, (according to the microscopic and mineral processing data) the optimum grain size for the beneficiation of allanite is the size fraction (10-212 μm).
Keywords: *Cerium, Lanthanum, Allanite, Crete.*

Περίληψη

Εντός των μεταμορφωμένων πετρωμάτων της Ομάδας των Πλακωδών Ασβεστολίθων των Ταλαίων Ορέων της κεντρικής Κρήτης εμφανίζονται ελαφρές σπάνιες γαίες (δημήτριο και λανθάνιο). Τα στοιχεία των σπανίων γαιών περιέχονται σε ένα ορυκτό της ομάδας του επιδότου το οποίο ονομάζεται αλλανίτης. Κατά την εξέταση μέσω του ηλεκτρονικού μικροσκοπίου σάρωσης εντοπίστηκε ο αλλανίτης (προκινηματικοί κρύσταλλοι) και καταγράφηκε η κοκκομετρία του. Στη συνέχεια, πραγματοποιήθηκαν δοκιμές εμπλουτισμού για τα επιλεγμένα δείγματα στα κοκκομετρικά κλάσματα (<10 μm), (10-212 μm) και (212-1000 μm), σύμφωνα με τις μαγνητικές ιδιότητες και το μέγεθος των κόκκων του αλλανίτη. Μετά από μαγνητικό διαχωρισμό των κλασμάτων (<10 μm) και (212-1000 μm) φάνηκε ότι οι σπάνιες γαίες (Ce, La) παρουσιάζουν υψηλότερες συγκεντρώσεις στα μαγνητικά προϊόντα. Από τις μικροσκοπικές παρατηρήσεις καθώς και τις δοκιμές εμπλουτισμού πρόεκυψε ότι η ιδανική κοκκομετρία εμπλουτισμού του αλλανίτη είναι (10-212 μm).
Λέξεις κλειδιά: *Δημήτριο, Λανθάνιο, Αλλανίτης, Κρήτη.*

1. Introduction

Rare earth metals are used in the modern industry in numerous technological fields, such as catalysts, electronics, glasswork, ceramics, special alloys and superconductors. Recently, the application of rare earths has focused on various research purposes, like geochronology and geochemical mapping, which is considered to play an important role in environmental research, geological mapping and mineral resources research. In addition, rare earths are used as geochemical tracers for hydrological, hydrogeological and oceanographic researches.

Recently, accessory LREE (Light Rare Earth Elements) bearing minerals have been investigated in eastern Crete, Greece, in a sequence of Palaeozoic amphibolite facies metapelites which were overprinted by Alpine low-T metamorphism (~300 °C). The series is dominated by Al, Ca rich metapelite layers, which contain Palaeozoic allanite, secondary Alpine monazite with low Y contents (<0.6 wt.%) or rhabdophane (Krenn and Finger, 2007). LREE-bearing minerals are also found within the metamorphic marine siliciclastic of Sisses and Fodele beds of the Plattenkalk Group (Manutsoglu, 2008). Mineral processing was conducted for the size fractions: (<10 μm), (10-212 μm) and (212-1000 μm), according to the magnetic properties and grain size of allanite.

Table 1 - Occurrences of allanite in Greece (updated until 2009).

Occurrence of allanite	Location	Author
Accessory mineral in amphibolites	Samos	Mezger and Okrusch (1985)
In andesitic rocks	Poros, NE Peloponnese	Mitropoulos P (1987)
Accessory mineral in Miocene I-type granitoids	Cyclades	Henjes-Kunst *et al.* (1988)
Accessory mineral in migmatites	Central Rhodope	Kotopouli *et al.* (1991)
In skarn formation	Serifos, Cyclades	Perdikatsis and Papastavrou (1993)
In granitoid rocks	Thessaly	Kotopouli *et al.* (2000)
Accessory mineral in Augen migmatite	Naxos, Cyclades	Keay *et al.* (2001)
Accessory mineral in quartz diorite	Delos, Cyclades	Pe-Piper *et al.* (2002)
In metamorphic rocks	Ikaria, Cyclades	Iliopoulos I (2005)
In magmatic epidote of the Sithonia pluton	Sithonia, Chalkidiki	Keramidas *et al.* (2005)
In phengite-rich eclogite	Syros, Cyclades	Putlitz *et al.* (2005)
In granodiorite	Tinos, Cyclades	Mastrakas N (2006)
Accessory mineral in tonalites and granodiorites	Sithonia, Chalkidiki	Christofides *et al.* (2007)
Accessory mineral in metapelites	Eastern Crete	Krenn and Finger (2007)
In metapelites and metasandstones	Central Crete	Manutsoglu E (2008)
Accessory mineral in mélange	Syros, Cyclades	Miller *et al.* (2009)
Accessory mineral in granites	Fanos, Northern Greece	Šarić *et al.* (2009)
In metapelitic garnets	Rhodope, Northern Greece	Mposkos *et al.* (2009)

The rare earths studied are seen in an epidote group mineral called allanite. Allanite occurs in metamorphic rocks between Sisses and Fodele beds of the Plattenkalk Group. There have been several references since the middle 80's about the allanite occurrences in Greece. Allanite occurs mainly as an accessory mineral in a variety of geological environments. Most of the studies are located at Cyclades (Aegean region). Allanite is found in skarn at Serifos, in migmatites at Naxos, in plutonic rocks at Delos, in eclogites and mélange rocks at Syros, in granodiorite at Tinos. Other occurrences of allanite are reported in metamorphic rocks from Ikaria, in amphibolites from Samos, in andesitic rocks from Poros, in metapelites from East Crete and in some areas in Northern Greece (in plutonite from Chalkidiki, in migmatites and gneiss from Rhodope, in granites from Fanos) and in granitoids from

Thessalia (Table 1). The four-year (2009-2013) ProMine project (Arvanitidis and Goodenough, 2014) has provided a well documented knowledge base of Europe's non-energy raw material resource potential, which is completed by various research outcomes regarding LREE distribution patterns (e.g. Melfos and Voudouris, 2012 and Papadopoulos *et al.*, 2015).

2. Geological settings

The Plattenkalk Group (Permian-Oligocene) represents the lowermost tectonic unit under a nappe pile of metamorphic and non metamorphic units of the External Hellenides and has undergone a prograde metamorphism ranging from anchimetamorphic to high pressure/low temperature facies conditions (Manutsoglu, 1990). The generally acceptable aspect for their depositional conditions has been derived from lithological comparisons with the succession of the Ionian Zone. It represents a transition from very shallow to deeper pelagic sedimentation. The older part of the Plattenkalk Group includes marine siliciclastic sediments and carbonates of late Permian to early Triassic age. These rocks are overlain conformably by a thick sequence of neritic carbonates with stromatolites of Upper Triassic-Liassic age. The next level comprises the sequence of platy limestones with bedded and nodular chert of Jurassic-Eocene age, which corresponds to the «Posidonia Schist» and «Viglaes facies» of Ionian zone. The uppermost stratigraphic level includes the transitional beds to a siliciclastic succession known as «Vathia Beds» on Peloponnes and “Kalavros Beds” on Crete, of Upper Eocene-Oligocene age. The Plattenkalk Group has been metamorphosed during Oligocene-Miocene (Manutsoglu, 1990). The discovery of extended colonies of lithistid demosponges in the middle parts (Jurassic-Eocene) of the chert-bearing metamorphic platy limestones from the Taygetos Mountains on the Peloponnes to the Ida Mountains on Crete allows the deduction of the sedimentary depositional conditions (Manutsoglu, *et al.* 1995; Soujon *et al.*, 1995 and Manutsoglu *et al.*, 1998, 2003).

3. Materials and Methods

Five samples were collected from the study area (Figure 1), two metasandstones and three schists (calc-schist, yellow and white schist). Mineralogical study through powder X-Ray diffraction (XRD) of these samples showed no evidence of allanite (Table 2). Thin section microscopy study showed microcrystals of allanite within crystals of albite. Scanning electron microscopy of the samples showed clearly the crystals of allanite (pre-kinematic crystals) and their grain size (approximately 30-100 μm) (Figure 2).

Table 2 - X-Ray Diffraction investigation of the initial samples.

Sample \ Mineral	Quartz	Albite	Muscovite	Calcite	Rutile	Hematite	Clino-chlore	Kaolinite
Metasandstone (1)	++	+	+	-	-	+	-	-
Metasandstone (2)	++	+	+	-	+	-	-	-
Schist (calc-schist)	+	-	+	++	+	-	+	-
Schist (yellow)	++	+	+	-	+	-	-	-
Schist (white)	++	+	++	+	-	-	-	+

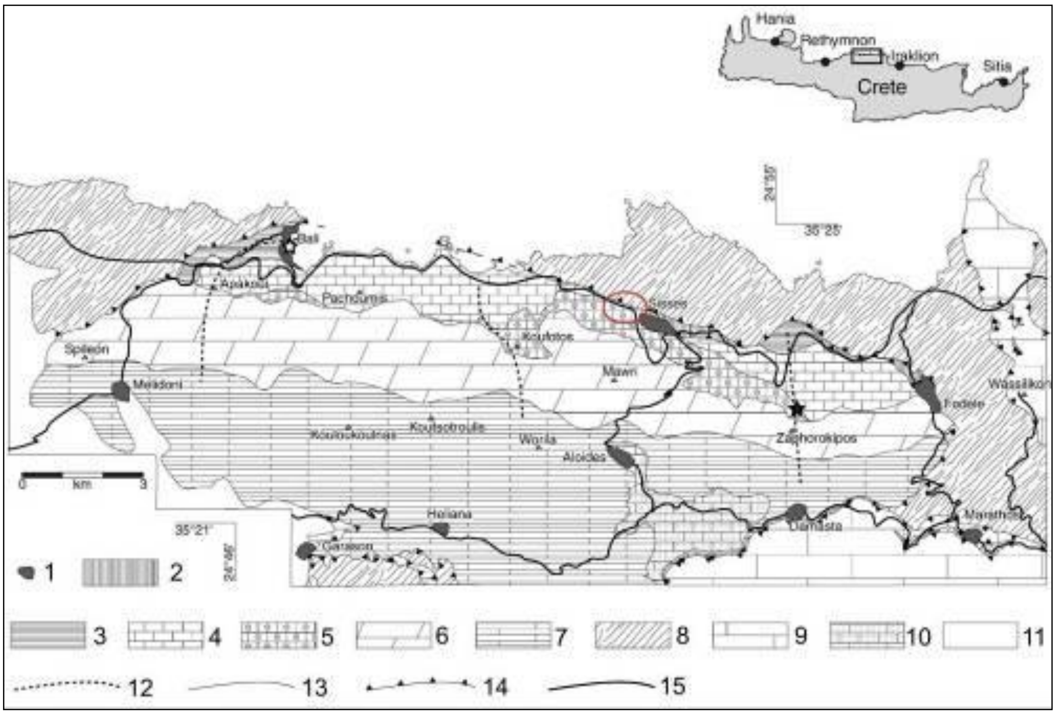

Figure 1 - Geological map of Talaia Ori (after Kock *et al*., 2007; modified by Epting *et al*., 1972). (1) Cities (2) Tectonized blocks (slivers), (3) Galinos beds (4) Fodele beds (5) Sisses beds (6) stromatolites-dolomites (7) Plattenkalk group (8) Phyllite-Quartzite nappe (9) Vasilikos marbles (10) red sediments (pre-Tortonio), (11) Tortonio-Pliocene (12) Trace of the stratigraphic section (13) Stratigraphic contact (14) Thrust fault (15) Roads.

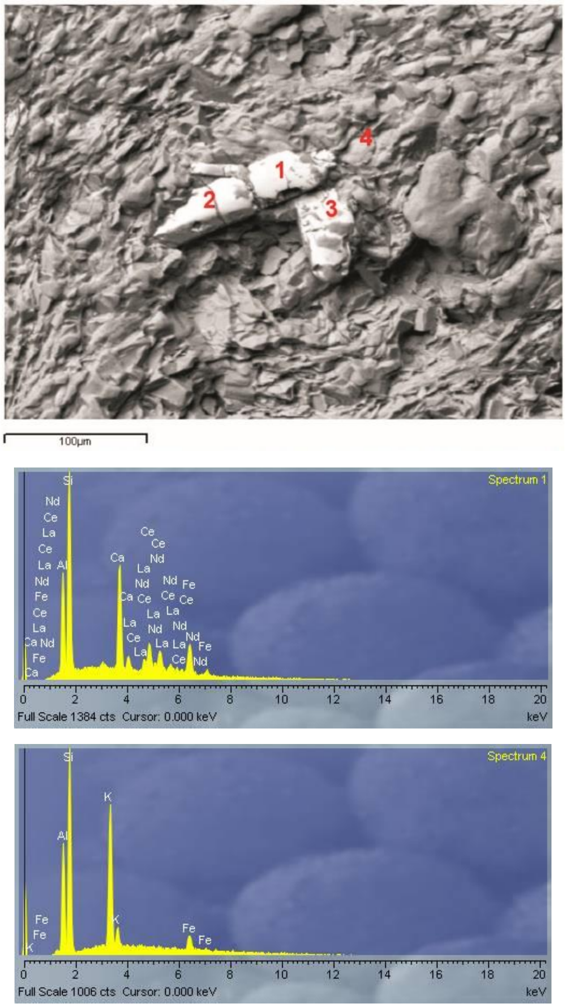

Figure 2 - Backscattered Electron (BSE) image of allanite (1) and mica minerals (4).

4. Sample preparation - Results

The original five samples (two metasandstones and three schists) went through crushing (using a jaw crusher) and then grinding in order to reduce their particle size. Next, each sample was homogenised. Using the Mozley hydrocyclon (2"), each homogenized sample was classified in two fractions <10 μm and >10 μm. The underflow (>10 μm) and overflow (<10 μm) of the hydrocyclon were collected separately and dried. Dry-sieving was carried out in the underflow in order to obtain the fractions (10-212 μm) and (212-1000 μm).

Table 3 includes weight (%) and concentration in Ce, La of each sample after hydroclassification and dry sieving.

4.1. Magnetic Separation - Results

Magnetic separation was conducted in the five original samples for the size fractions (<10 μm), (10-212 μm) and (212-1000 μm). The fine fraction (<10 μm) was separated using the wet high intensity magnetic separator (Carpco), whilst the fractions (10-212 μm) and (212-1000 μm) were separated using the high intensity induced roll magnetic separator (MIH 111-5, Carpco). The results for each fraction are shown in Tables 4, 5 and 6.

Table 3 - Results after hydroclassification and dry sieving.

Sample	Size fraction (µm)	Weight (%)	La (ppm)	Ce (ppm)
Metasandstone (1)	212-1000	83,48	7,95	14,56
	10-212	7,88	6,87	8,39
	<10	8,64	44,94	92,12
	Total	100,00	11,90	22,96
Metasandstone (2)	212-1000	77,61	8,68	16,52
	10-212	6,22	8,29	13,48
	<10	16,17	26,18	47,88
	Total	100,00	10,92	19,60
Schist (calc-schist)	212-1000	53,26	15,12	23,80
	10-212	21,27	17,08	25,62
	<10	25,47	13,38	35,00
	Total	100,00	11,34	23,80
Schist (yellow)	212-1000	61,92	33,32	58,10
	10-212	12,84	8,83	17,36
	<10	25,24	61,04	106,68
	Total	100,00	25,20	50,40
Schist (white)	212-1000	59,14	34,44	82,32
	10-212	15,13	46,06	104,30
	<10	25,73	12,91	47,46
	Total	100,00	37,66	84,74

Table 4 - Magnetic separation results for the fine fraction (<10 µm).

Sample	Product	Weight (%)	La (ppm)	Ce (ppm)
Metasandstone (1)	**Magnetic**	14,38	49,70	107,80
	Non magnetic	85,62	42,98	88,76
	Total	100,00	44,94	92,12
Metasandstone (2)	**Magnetic**	7,03	39,06	94,08
	Non magnetic	92,97	21,00	49,42
	Total	100,00	26,18	47,88
Schist (calc-schist)	**Magnetic**	2,61	45,08	138,18
	Non magnetic	97,39	13,80	48,30
	Total	100,00	13,38	35,00
Schist (yellow)	**Magnetic**	6,63	54,88	105,56
	Non magnetic	93,37	58,80	119,56
	Total	100,00	61,04	106,68
Schist (white)	**Magnetic**	1,53	152,60	358,40
	Non magnetic	98,47	9,49	37,80
	Total	100,00	12,91	47,46

Table 5 - Magnetic separation results for the fraction (10-212 µm).

Sample	Product	Weight (%)	La (ppm)	Ce (ppm)
Metasandstone (1)	**Magnetic**	20,73	14,36	35,91
	Intermediate	18,91	4,94	10,28
	Non magnetic	60,36	5,10	5,18
	Total	100,00	6,87	8,39
Metasandstone (2)	**Magnetic**	36,08	13,72	26,68
	Intermediate	19,80	7,36	8,85
	Non magnetic	44,12	4,05	6,99
	Total	100,00	8,29	13,48
Schist (calc-schist)	**Magnetic**	17,05	27,60	54,04
	Intermediate	28,73	15,54	25,90
	Non magnetic	54,22	11,37	18,20
	Total	100,00	17,08	25,62
Schist (yellow)	**Magnetic**	44,33	39,74	80,31
	Intermediate	17,17	18,90	33,32
	Non magnetic	38,50	12,75	21,70
	Total	100,00	8,83	17,36
Schist (white)	**Magnetic**	21,84	108,23	230,85
	Intermediate	21,71	39,20	93,66
	Non magnetic	56,45	28,42	62,86
	Total	100,00	46,06	104,30

Table 6 - Magnetic separation results for the fraction (212-1000 μm).

Sample	Product	Weight (%)	La (ppm)	Ce (ppm)
Metasandstone (1)	**Magnetic**	8,50	6,35	17,66
	Intermediate	15,50	4,56	7,90
	Non magnetic	76,00	5,25	7,55
	Total	100,00	7,95	14,56
Metasandstone (2)	**Magnetic**	19,22	10,07	17,35
	Intermediate	21,17	3,72	12,45
	Non magnetic	59,61	5,66	9,98
	Total	100,00	8,68	16,52
Schist (calc-schist)	**Magnetic**	2,29	28,64	71,72
	Intermediate	7,92	17,36	24,50
	Non magnetic	89,79	12,56	18,06
	Total	100,00	15,12	23,80
Schist (yellow)	**Magnetic**	20,76	33,44	71,78
	Intermediate	19,11	31,78	65,66
	Non magnetic	60,13	24,92	51,94
	Total	100,00	33,32	58,10
Schist (white)	**Magnetic**	16,80	73,03	165,34
	Intermediate	15,88	33,46	70,00
	Non magnetic	67,32	34,16	70,00
	Total	100,00	34,44	82,32

5. Conclusions

The abundance of allanite in the Sisses Beds, strengthens the paleobathymetric/depositional model, which is proposed from Dornsiepen *et al.* (2001) according to which, the Permian Triassic sequence of the Plattenkalk Group was fine grained, well sorted siliciclastics of a marine deltaic environment, on the southern margin of the Palaeothethys belonging to Gondwana.

Metasandstone (1), metasandstone (2) and yellow schist samples show higher rare earth (Ce, La) concentrations in the fraction (<10 μm). Lower rare earth concentrations were found in the fraction (10-212 μm) of the metasandstone (1) and the yellow schist. The calc-schist sample does not seem to have notable rare earth concentrations in any fraction. The white schist sample has higher rare earth concentrations in the fraction (10-212 μm), and lower concentrations in the fractions (212-1000 μm) and (<10 μm).

The magnetic separation of the fraction (<10 μm) showed that the rare earth concentrations are higher in the magnetic products of all the initial samples, except from the yellow schist.

In the fraction (10-212 μm) rare earth concentrations are clearly higher in the magnetic products of all samples.

On the other hand, rare earth concentrations in the fraction (212-1000 μm) are higher in the magnetic products of metasandstone (1), metasandstone (2) calc schist and white schist, whereas there are no notable concentrations in the yellow schist separation products.

In conclusion, (according to the microscopic and mineral processing data) the optimum grain size for the beneficiation of allanite is (10-212 μm). A systematic petrologic study for the understanding of the origin/formation of allanite within the particular metamorphic rocks of the Plattenkalk Group would also provide useful results to optimize the mineral processing methodology.

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