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FORMATION AND COMPARISON OF RARE EARTH ELEMENT (REE) GEOCHEMISTRY OF MALATYA FLUORITE DEPOSITS

MALATYA FLORİT YATAKLARININ OLUŞUMU VE NADİR TOPRAK ELEMENT (NTE) JEOKİMYASININ KARŞILAŞTIRILMASI

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ABSTRACT

The fluorite deposits in Malatya (Turkey) region are observed in Yeşilyurt and Kuluncak districts. Yesilyurt fluorites are observed as vein type along the plane of unconformity between metamorphic rocks on hydrothermal phase. Kuluncak fluorites are observed in the contacts of alkaline syenites and marbles as vein type which the mineralization occured in the pegmatitic phase as shown on Tb/La-Tb/Ca diagram. Yeşilyurt fluorites are seen in purple, dark purple and blackish colors and Kuluncak is blue to purple. When the geochemical characteristics of the Malatya fluorites are taken into comparison, the Yeşilyurt region has 12.2-35.8% F values and Kuluncak has 15, 03-25, 42%. The REE contents of Kuluncak fluorites are richer than Yeşilyurt fluorites as well as in Sr contents. In the $(La/Yb)_N$-$(Tb/Yb)_N$ diagram the Yeşilyurt fall into the field of New Mexico, however the Kuluncak fluorites fall into the field of Akdağmadeni. The fluorite deposites are compared on $(Eu/Eu^*)_n$–$(La/Yb)_n$ and Sc–ΣREE, $(Tb/Yb)_n$–$(La/Yb)_n$ diagrams. The Yeşilyurt fluorites display a distribution similar to the Buyukkizilcik (Kahramanmaras) fluorites but Kuluncak fluorites spread in large areas. The normalized-REE compositions of the Malatya fluorites exhibit enrichment in LREEs compared to the HREEs. In the light of all the data obtained, it can be concluded that the geochemical properties and formation of Malatya fluorites are different from each other.

Keywords: Fluorite, Hydrothermal, Pegmatitic phase, REE, Malatya.

ÖZET

Malatya'daki (Türkiye) florit yatakları Yeşilyurt ve Kuluncak bölgelerinde bulunmaktadır. Yeşilyurt floritleri, hidrotermal evrede metamorfik kayaçlar arasındaki uyumsuzluk düzlemi boyunca damar tipi olarak görülmektedir. Kuluncak floritleri ise Tb/La-Tb/Ca diyagramında gösterildiği gibi, pegmatitik evrede oluşmakta ve alkali siyenit ve mermer dokanağında damar tipi şeklinde görülmektedir. Yeşilyurt floritleri mor, koyu mor ve siyahımsı renklerde, Kuluncak floritleri ise maviden mora kadar farklı renklerde görülmektedir. Malatya bölgesinde farklı lokasyonlarda gözlenen floritlerin jeokimyasal özellikleri karşılaştırıldığında; Yeşilyurt bölgesindeki F % 21,2-35,8 değerlerinde; Kuluncak bölgesinde ise F % 15,03- 25,42 arasındadır. Kuluncak floritlerinin nadir toprak element (NTE) ve Sr içeriği, Yeşilyurt floritlerininkinden daha zengindir. $(La/Yb)_N$-$(Tb/Yb)_N$ diyagramlarında Yeşilyurt floritleri New Mexico alanına düşerken, Kuluncak floritleri Akdağmadeni floritleri ile aynı alandadır. Florit yataklarının $(Eu/Eu^*)_n$–$(La/Yb)_n$ ve Sc–ΣNTE, $(Tb/Yb)_n$–$(La/Yb)_n$ diyagramlarında karşılaştırmaları yapılmıştır. Yeşilyurt floritleri Büyükkızılcık (Kahramanmaraş) floritlerine benzer bir dağılım sergilerken, Kuluncak floritleri geniş alanlara yayılmıştır. Malatya floritlerinin normalize edilmiş Nadir Toprak Element (NTE) bileşimlerine göre, hafif nadir toprak elementlerinin ağır nadir toprak elementlerine göre zenginleştiği görülmektedir. Elde edilen tüm veriler ışığında Malatya floritlerinin Jeokimyasal özellikleri ve oluşumlarının birbirlerinden farklı olduğu sonucuna varılabilir.

Anahtar Kelimeler: Florit, Hidrotermal, Pegmatitik evre, NTE, Malatya.

INTRODUCTION

Ketin (1966) classified the orogenic belts of Turkey into 4 different classes as Pontides, Anatolides, Taurides and Border folds. In Malatya (Figure 1) where is the tectonic location on Eastern Taurus Orogenic region, Doğanşehir, Malatya, Doğanyol and Pütürge consists of Precambrian-Paleozoic and Mesozoic aged gneiss, amphibolite, marble and schists (MTA, 2009). Miocene-Pliocene aged volcanic rocks are located in the north of Hekimhan-Arguvan (Atabey, 2015). Upper Cretaceous ophiolites, Cretaceous limestones, Eocene volcanoclastic and sedimentary rocks and Miocene-Pliocene lacustrine, evaporitic sedimentary rocks are located in different locations (Perinçek and Kozlu, 1984; Atabey, 2015). The fluorite veins which developed in Malatya region have been studied by Revan and Genç (2003) in Yeşilyurt and Özgenç and Kibici (1994). Altuncu (2009) stated that the Yeşilyurt fluorites are poor in terms of REE and the trace element association is in the form of F+Ba+Si+Au. Kuluncak stated that the fluorite bed is formed as vein fillings and irregular veins in contact with syenite limestone due to alkali magmatism in the region. It also states that the formation of fluorite forms at the pegmatitic stage (Altuncu, 2009). Depending on the type of rare earth element-rich rocks fluorite deposits in Turkey are divided into three classes: (i) carbonatite, (ii) alkaline igneous rocks, and (iii) limestone (Altuncu, 2009). Öztürk et al. (2019) reported that these deposits developed in relation to post-collision magmatism and that ore-forming fluids were fed from other sources.The aim of this study is comparing the formation and REE geochemistry of Malatya fluorites.

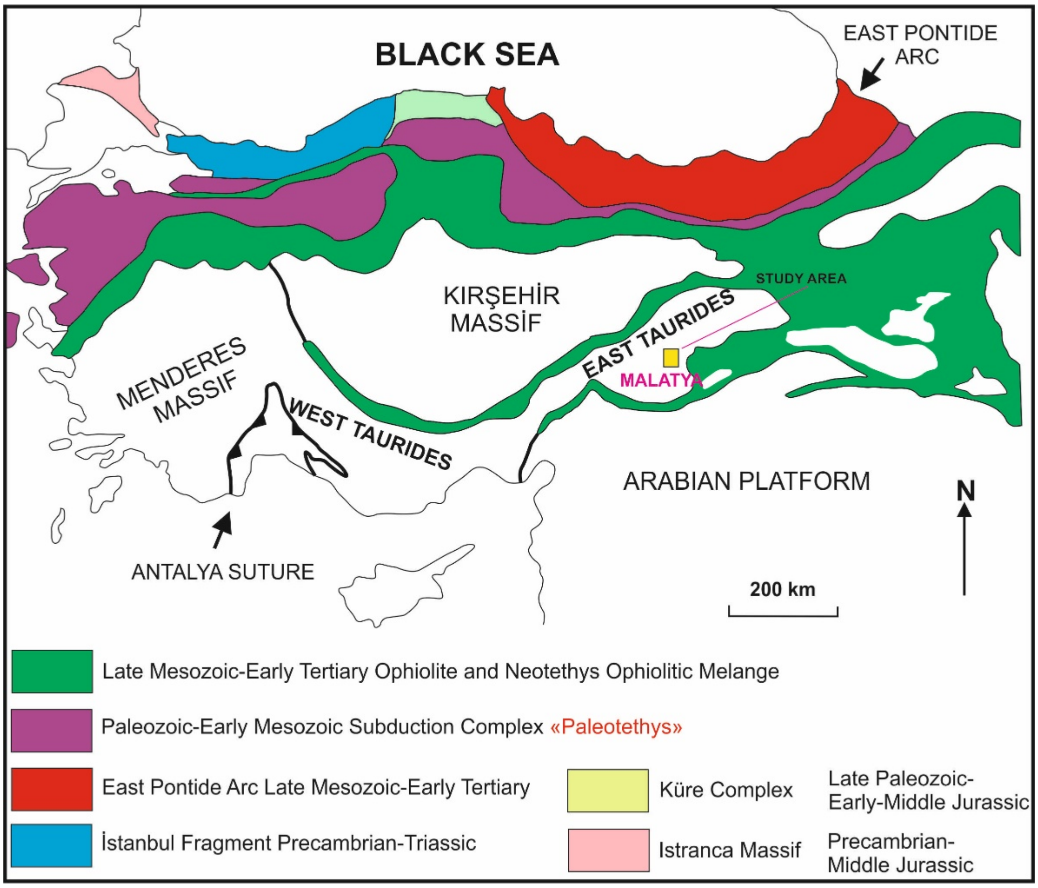

Figure 1. Location of the study area on the tectonic units of Turkey (Quoted as modified from Şengör et al., 1982)

Geological Settings

The Yeşilyurt fluorites were formed (Fig 2) along the discordance plane between the Devonian-Carboniferous aged Kalecik marbles and the Permian aged Düzağaç schists are located at the west and south of Kuz Tepe (Fig 2) (Revan, 2003). Mineralized zone has a geometry which is parallel to the discordance plane and which continues by thinning towards the sides (Fig 3). Faults with NW-SE orientation are generally observed in the area of examination. It is considered that these broken faults, which are observed systematically, are effective in mineralization. Fluorite mineralization occurred as the hot waters circulating around the fault lines transferred the mineralized solutions and discharged those into suitable areas. Observation of mineralization in the schist and limestone discordance plane occurred as the solutions moving in faults and cracks encountered impermeable schists and left their solutions in this discordance plane.

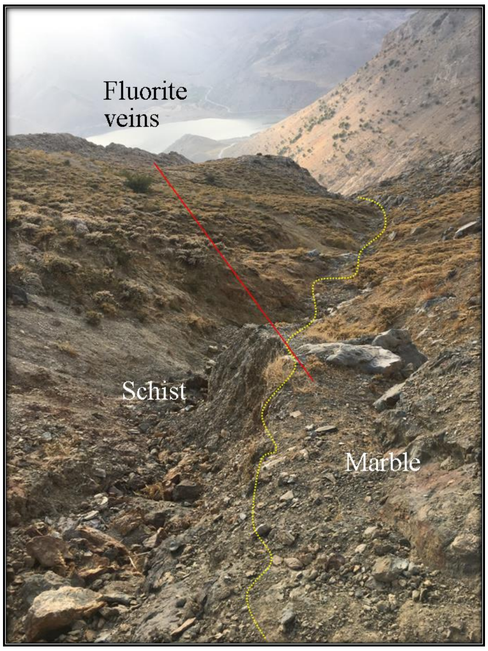

Figure 2. Location of fluorite mineralization on western part of Kuz Tepe

The Kuluncak fluorites are related to the syenite-limestone contact (Fig 4) in the region (Özgenç, 1999). Altuncu (2009) stated that the Kuluncak fluorite deposit is formed as vein fillings and irregular veins in contact with syenite limestone due to alkali magmatism in the region. In fluid inclusion studies Altuncu (2009) reported that the homogenization temperature (Th) is above 580°C on average. Fluorite mineralization in the study area is located in Aşılık Pınarı, Ardıçlı Tepe and Alibeyli (Fig 5).

The Yeşilyurt Fluorites are observed in purple, light purple and blackish colours (Fig 6) but the Kuluncak fluorites are purple (Fig 7).

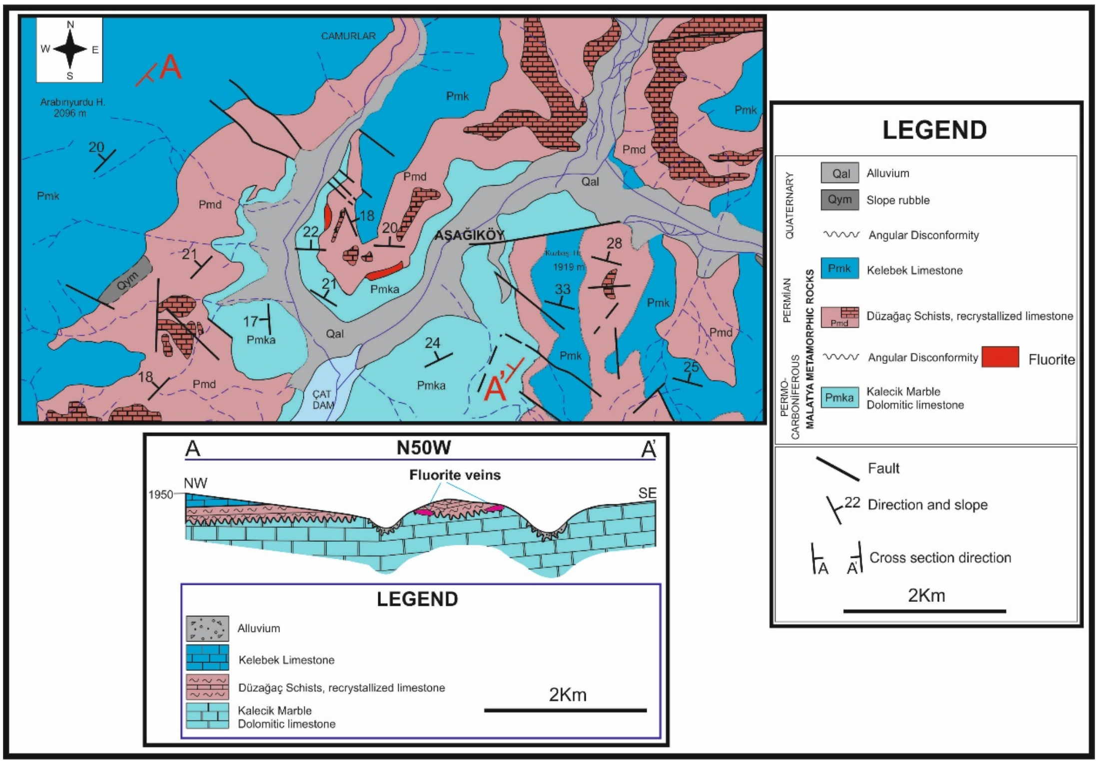

Figure 3. Geological map of the Aşağıköy (Yeşilyurt-Malatya) area and the geological cross section with NW-SE orientation (Modified from Revan, 2003)

Figure 4. General view of quarries with fluorite mineralizations

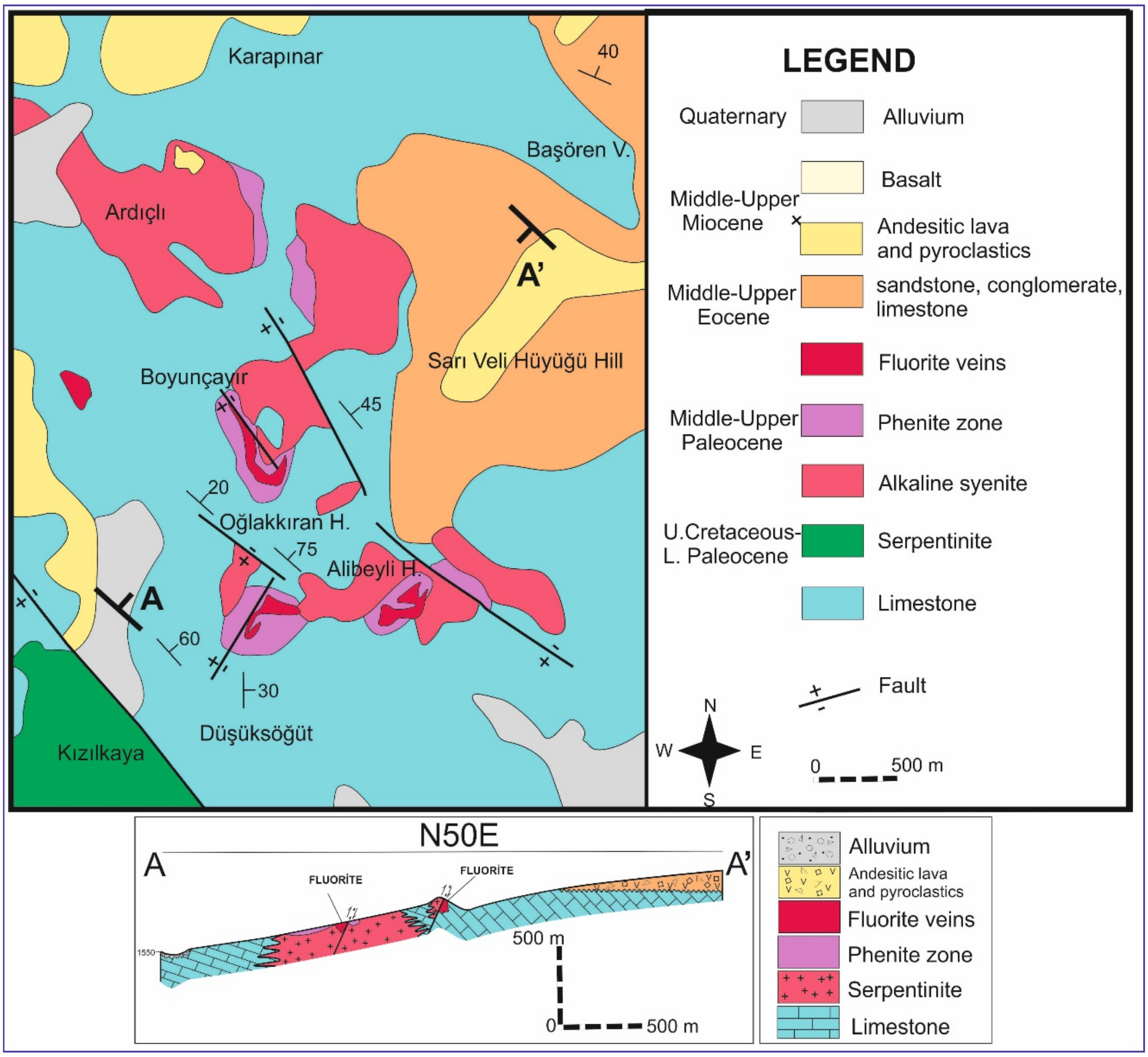

Figure 5. Geological map of the Kuluncak area and the geological cross section with NE-SW orientation (Modified from Özgenç, 1999)

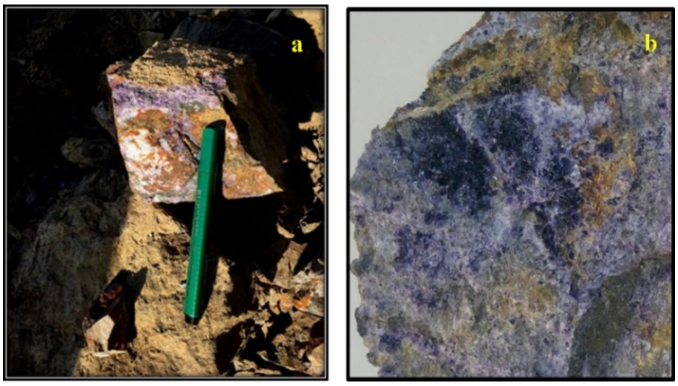

Figure 6. Appearances of purple fluorites in the Yeşilyurt district

Figure 7. Appearances of purple fluorites in the Kuluncak district

METHOD

Rare earth elements are used as a geological indicator in order to reveal the genesis of rocks and ore deposits. The determination of the geotectonic environment of magmatic melts is of importance in terms of revealing the origins and learning the data related to the crystallization conditions. The fluorite samples taken from Yeşilyurt and Kuluncak (Malatya) region were fractured in the geochemistry laboratory of Kahramanmaraş Sütçü İmam University and separated by their colours in the form of pure crystals. The fluorite samples were sorted separately as pure fluorite and decomposed fluorite. Selected crystals are grounded in agate mortar, packaged as 5-grams 20 samples were sent to Acme Laboratories (Vancouver–Canada) and their rare earth element analyses are performed using $LİBO_2$ fusion, ICP-MS method.

RESULTS AND DISCUSSION

REE Geochemistry

The rare earth element contents of the Yeşilyurt fluorites present a poor content which are similar contents were observed in Feke fluorites (Uras, 2002) and in Büyükkızılcık fluorites (Uras and Çalışkan, 2014) but Kuluncak fluorites present a very rich content also Sr % F values change between 12.2-35.8 (Table 1) in Yeşilyurt and 15,03-25,42 (Table 2) in Kuluncak district. The rare earth element contents of Yeşilyurt fluorites range from 0.02 to 25.8 ppm (Table 1) and the rare earth element contents of Kuluncak fluorites vary between 0.11 and 789.5 ppm (Table 2). A total of 10 fluorite samples contain 369,64 ppm of rare earth elements in Yeşilyurt (Table 1). Total amount of rare earth elements of Kuluncak fluorites varies between 167.01-1188.82 ppm (Table 2).

Terbium (Tb) and Lanthanum (La) are subjected to extensive disaggregation during the formation of fluorite. For this reason, the formation environment and the degree of disaggregation of the mineral are determined by evaluating Tb/La and Tb/Ca ratios (Schneider et al. 1975, Möller et al. 1976; Möller and Morteani, 1983). The Yeşilyurt fluorites fall in the hydrothermal origin field and Kuluncak in the pegmatitic origin field in the Tb/Ca vs. Tb/La diagram (Figure 8).

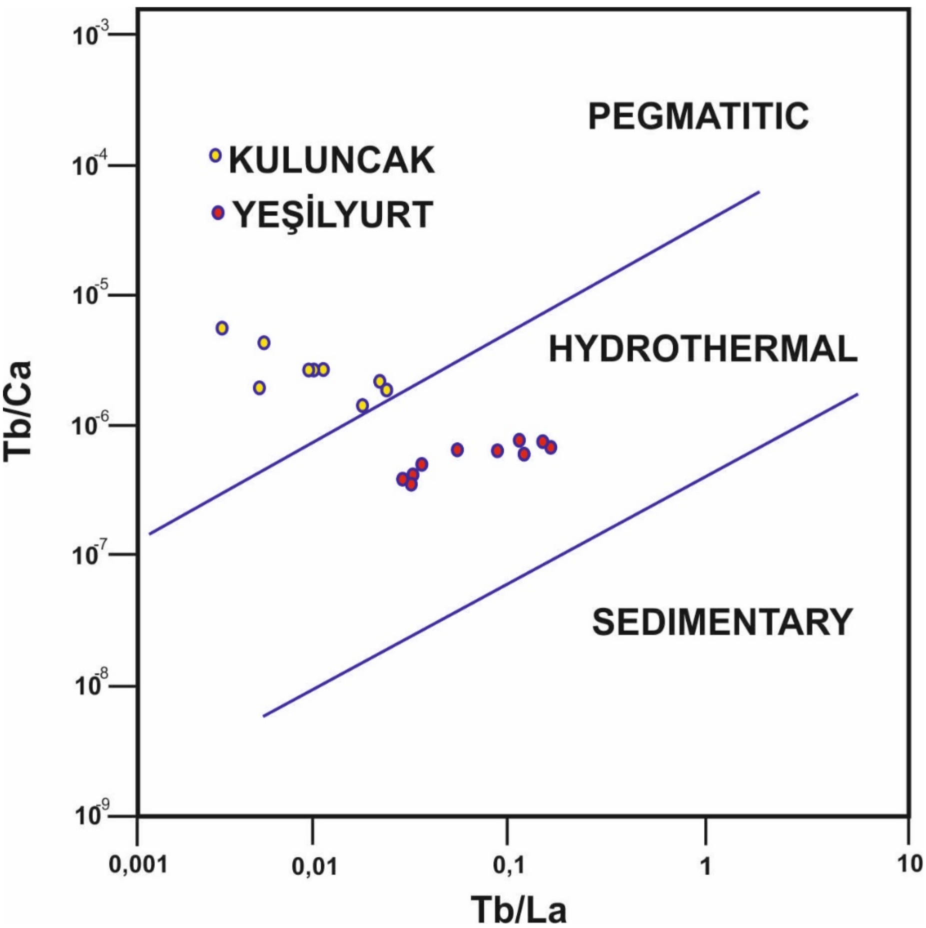

Figure 8. The place of the fluorites in the Tb/La vs. Tb/Ca diagram (Möller and Morteani, 1983)

Table 1. % F, Sr and rare earth element contents of the Yeşilyurt fluorites

Sr (ppm)	37,6	36,4	31,3	32,3	24,2	37,0	38,4	38,8	39,9	35,8
F%	21,69	12,2	13,05	14,09	18,8	35,8	20,00	19,88	20,5	19,78
La (ppm)	3,4	5,1	4,6	4,2	4,9	5,3	2,8	2,6	3,7	2,3
Ce (ppm)	6,0	8,2	6,0	7,0	7,5	5,1	5,3	6,4	5,6	6,2
Pr (ppm)	0,97	1,12	0,91	0,82	1,09	0,84	0,85	0,90	0,90	0,84
Nd (ppm)	4,6	4,5	3,8	3,4	3,2	3,9	4,3	4,3	4,3	3,2
Sm (ppm)	1,26	0,84	0,72	0,82	0,71	1,11	1,11	1,29	1,06	1,27
Eu (ppm)	0,32	0,18	0,14	0,12	0,14	0,24	0,27	0,34	0,31	0,31
Gd (ppm)	1,85	0,90	0,75	0,82	0,78	1,44	1,64	1,74	1,53	1,58
Tb (ppm)	0,32	0,15	0,12	0,11	0,12	0,24	0,28	0,33	0,27	0,31
Dy (ppm)	2,02	0,91	0,79	0,64	0,74	1,47	1,60	2,16	1,69	2,12
Ho (ppm)	0,40	0,20	0,18	0,14	0,18	0,21	0,39	0,44	0,38	0,41
Er (ppm)	1,07	0,59	0,38	0,28	0,45	0,71	0,99	1,16	0,95	1,15
Tm (ppm)	0,12	0,05	0,05	0,05	0,06	0,10	0,10	0,12	0,10	0,11
Yb (ppm)	0,66	0,32	0,24	0,34	0,31	0,49	0,58	0,62	0,61	0,59
Lu (ppm)	0,08	0,04	0,03	0,02	0,09	0,07	0,07	0,08	0,07	0,09
Sc (ppm)	1	1	0,1	0,1	0,1	0,1	0,1	0,1	0,1	0,1
Ce\Yb	9,09	25,63	25,00	20,59	24,19	10,41	9,14	10,32	9,18	10,51
Tb \La	0,094	0,029	0,026	0,026	0,024	0,045	0,100	0,127	0,073	0,135
Tb\Ca*10 6	0,0000007 57	0,0000004 97	0,0000003 95	0,0000003 71	0,0000003 89	0,0000006 54	0,0000006 17	0,0000007 30	0,0000006 47	0,0000006 97
Ce\Ce*	0,8	0,8	0,7	0,9	0,8	0,6	0,8	1,0	0,8	1,1
Eu/Eu*	0,6	0,6	0,5	0,4	0,5	0,5	0,6	0,6	0,7	0,6

ΣREE	44,67	32,30	27,11	26,56	28,57	40,22	41,98	45,68	39,97	42,58

The studied samples from Malatya fluorites cover the crystallized region put forward by Hill et al. (2000) in $(Tb/Yb)_N$ vs. $(La/Yb)_N$ diagram. The Yeşilyurt fluorites are located in the region where the New Mexico fluorites are located but Kuluncak fluorites are located in Akdagmadeni area (Figure 9).

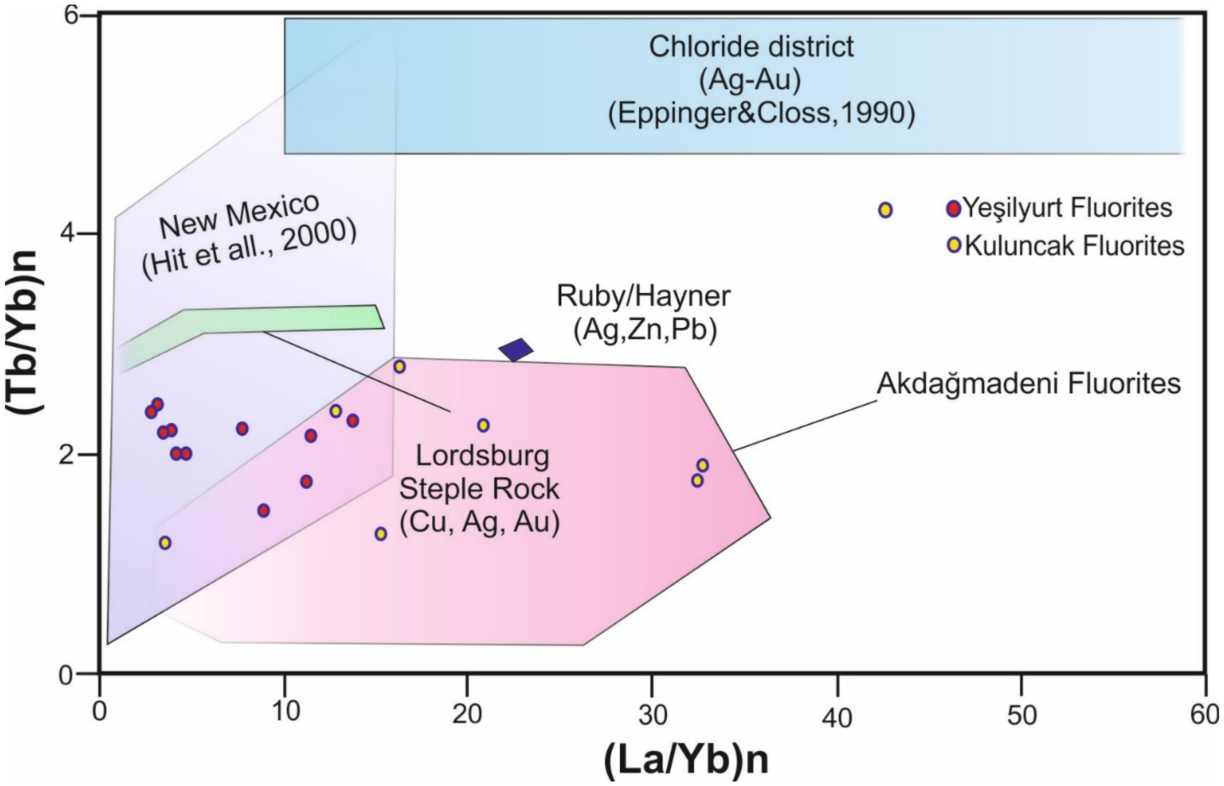

Figure 9. Locations of the studied fluorites in the $(Tb/Yb)_N$ vs. $(La/Yb)_N$ diagram (all values are normalized by comparing (Tb/Yb)n and (La/Yb) The n ratios in Malatya fluorites (Eppinger and Closs, 1990; Hill et al, 2000; Şaşmaz et al, 2005)

Looking at the place of Malatya fluorites within the diagram obtained by comparing the REE results, (La/Yb)n and (Eu/Eu)n ratios, it is observed that Yeşilyurt are located in the same region with the Büyükkızılcık fluorites (Uras and Calışkan, 2014) but Kuluncak fluorites spread in large areas (Fig 10).

Table 2. % F, Sr and rare earth element contents of the Kuluncak fluorites

Sr (ppm)	1432,4	1700,1	1648,8	1781,5	1407,9	2024,9	1603,7	2038,9	1676,4	1485,5
F%	19,33	25,2	25,39	17,1	15,03	17,34	19,37	25,42	18,25	19,86
La (ppm)	105,7	45,4	46,7	380,6	758,7	125,0	43,3	216,8	144,6	138,2
Ce (ppm)	157,6	53,1	56,0	463,7	789,5	178,4	80,9	255,6	203,2	211,1
Pr (ppm)	15,31	6,15	6,19	34,90	56,09	14,46	5,96	18,88	17,29	16,21
Nd (ppm)	46,5	19,8	20,5	91,1	142,6	39,4	17,6	54,1	53,0	50,1
Sm (ppm)	7,64	4,08	3,82	12,18	14,67	5,41	3,41	6,22	7,97	7,65
Eu (ppm)	1,62	0,94	0,89	2,30	2,67	1,28	0,71	1,13	1,54	1,25
Gd (ppm)	8,48	5,19	5,39	10,29	12,93	6,40	3,38	6,54	7,78	6,98
Tb (ppm)	1,41	0,87	0,87	1,68	2,06	1,12	0,64	0,92	1,12	1,09
Dy (ppm)	9,13	5,30	5,57	8,68	11,87	6,86	4,04	5,04	6,59	5,63
Ho (ppm)	1,94	1,24	1,34	1,90	2,65	1,95	0,90	1,31	1,55	1,35
Er (ppm)	5,10	3,42	3,64	5,43	7,30	5,38	3,36	3,84	3,79	3,52
Tm(ppm)	0,59	0,34	0,39	0,70	0,97	0,87	0,38	0,41	0,47	0,41
Yb (ppm)	2,80	1,39	1,63	3,15	5,29	3,98	2,42	2,85	2,81	2,62
Lu (ppm)	0,23	0,12	0,14	0,37	0,62	0,41	0,11	0,18	0,14	0,13
Sc (ppm)	1	0,1	0,1	0,1	0,1	0,1	0,1	0,1	0,1	0,1
Ce\Yb	56,29	38,20	34,36	147,21	149,24	44,82	33,43	89,68	72,31	80,57
Tb \La	0,013	0,019	0,019	0,004	0,003	0,009	0,015	0,004	0,008	0,008
Tb\Ca	0,000000312	0,000001773	0,000001827	0,000003886	0,000005058	0,000002453	0,000001298	0,000001812	0,000002380	0,000002448
Ce\Ce*	1,0	0,8	0,8	1,0	0,9	1,0	1,2	1,0	1,0	1,1

Eu/Eu*	0,61	0,62	0,60	0,62	0,59	0,66	0,64	0,54	0,59	0,52
ΣREE	354,35	194,34	201,27	739,98	1188,82	359,72	167,01	446,82	383,95	382,46

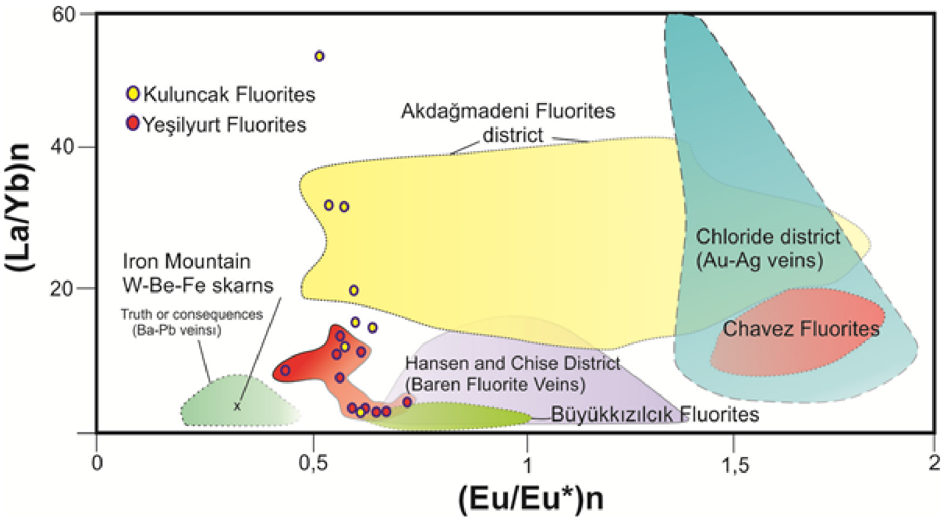

Figure 10. Locations of the Malatya fluorites in the (La/Yb)N vs. (Eu/Eu*)N diagram (Şaşmaz et al, 2005; Uras and Çalışkan, 2004)

In the Sc vs. ∑REE diagram (Şaşmaz et al, 2005), Yeşilyurt fluorites are distributed in a narrow area while Kuluncak fluorites are distributed in large areas (Fig 11).

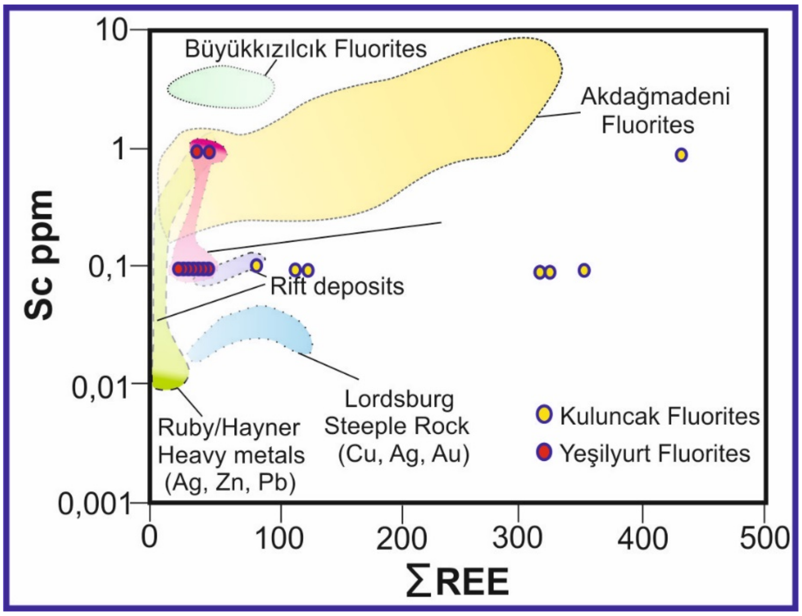

Figure 11. Comparison of Sc vs. ∑REE diagram (Şaşmaz et al, 2005)

The REE compositions of the Malatya fluorites were normalized to Chondrite values and show the light rare earth elements (LREE) are over-rich, as a consequence of continental crust effect, compared to the heavy rare earth elements (HREE) (Fig 12). In normalized diagrams, the positive Ce anomaly of fluorites indicates the presence of low oxygen fugacity at the source of hydrothermal fluids, while the negative Eu anomaly indicates that these conditions also persist in the mineralization environment (Möller and Morteani, 1983; Brookins, 1989).

In this study, rare earth element contents were evaluated by being normalized according to C1-Chondrites (Evensen et al, 1978). It is known that, in case the solutions circulating inside the rock are rich in carbonates, rare earth elements exhibit a more mobilized character compared to the normal aqueous solution (Rollinson, 1993). No significant crystallization has been observed in the normalized rare earth element diagrams of Malatya fluorite samples. With regard to Malatya fluorites, rare earth elements generally present a discordant behaviour during the magmatic crystallization and are enriched in the waste fluid. This presents an enrichment in minerals created in HREE late phase, whereas HREE with small radius also enter in the structure of the minerals created first. Therefore, LREE contents also present enrichment. While LREE contents provide a high value, HREE and LREE values present a lesser enrichment compared to LREE (Figure 12).

The Ce/Ce* and Eu/Eu* ratios provide important data on the oxygen content of the environment (Rollinson, 1993). According to Constantopoulos (1988), if the values of Eu/Eu* are >1, Eu^{+2} transforms into Eu^{+3} by being oxidized (as there is enough oxygen in the atmosphere) and enters into the lattice structures of fluorites. If the Ce/Ce* values are <1, this will cause the Ce to combine with oxygen and precipitate as CeO_2, thus preventing its entry into the structure of fluorites. It causes a negative anomaly to arise for Ce and a positive anomaly to arise for Eu in diagrams normalized with chondritic values (Evensen et al, 1978).

In Eu and Ce anomalies, the fact that Eu presents a negative anomaly and that Ce presents a positive anomaly indicates an environment with low temperature (T) and low oxygen fugacity ($_fO2$). In addition, the negative Eu anomaly is indicative of the presence of Eu^{+2} in the hydrothermal fluid. Ce/Ce* and Eu/Eu* ratios are calculated with $Ce/Ce^*=Ce_n/(La_n \times Pr_n)_{1/2}$ and $Eu/Eu^*=Eu_n/(Sm_n x Gd_n)_{1/2}$ formulas (Constantopoulos, 1988; Palmer and Williams-Jones, 1996; Williams-Jones et al., 2000). Yeşilyurt fluorites are formed under the influence of hydrothermal fluids. The fact that fluorites that are unaffected by a magmatic fluid are poor in terms of High Field-Strength Elements (HFSE) and rare earth elements (REE) indicates that these elements are affected by mineral forming solutions (Yaman, 1985).

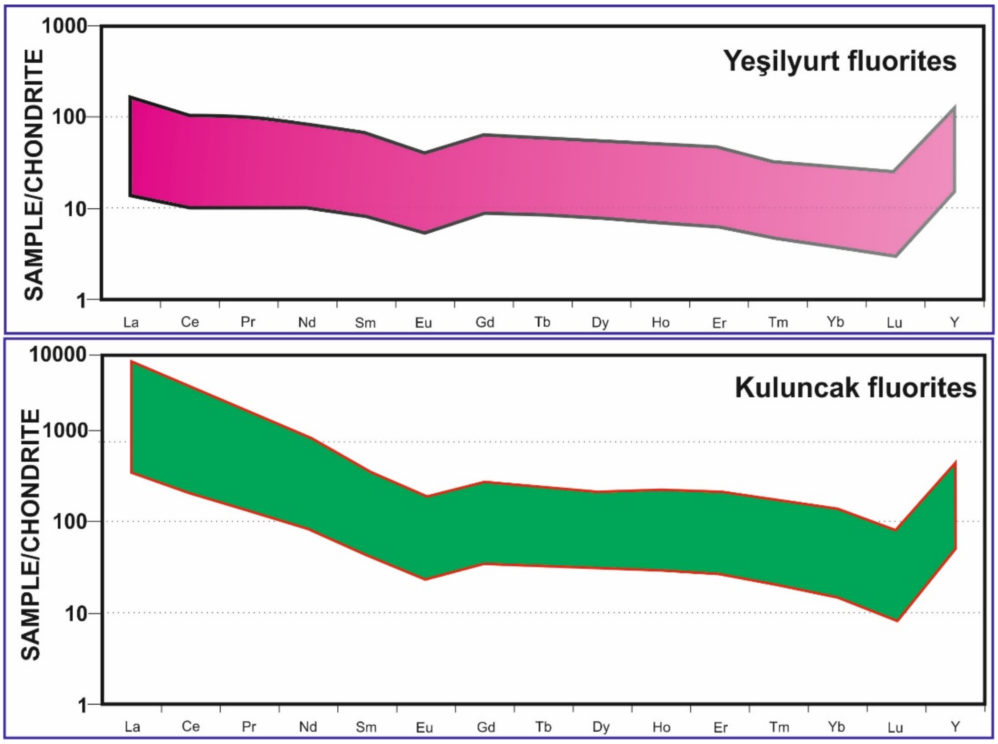

Figure 12. Chondrite- normalized REE spider diagram for (a) Yeşilyurt fluorites and (b) Kuluncak fluorites. (Rare earth element contents were evaluated by being normalized according to C1-Chondrites (Evensen et al, 1978))

DISCUSSION

This study reveals the REE chemical characteristics of Malatya fluorites located in the Eastern Taurus Orogenic belt. The REE contents of the Yeşilyurt fluorites present a poor content. Similar contents were observed in Feke fluorites (Uras, 2002), Büyükkızılcık fluorites (Uras and Çalışkan, 2014) and in Sarıveliler fluorites (Uras et al., 2019). But the REE contents of the Kuluncak fluorites present a rich content.

Altuncu (2009) has been investigated and compared the occurrence of fluorite mineralizations in Turkey and observed the formation environment and origin of fluorites on the Taurus orogenic belts. Coşanay et al. (2017)

demonstrated the microthermometric and geochemical properties of the fluorites observed in the Middle Anatolia. Azizi et al. (2018) examined the fluorites in the Markazi region in Iran and stated that vein type fluorites are poor in REE and dolomite, barite, hematite, goethite and small amount of pyrite are observed in paragenesis.

CONCLUSIONS

As a result; Malatya fluorites where vein type mineralization is seen on Eastern Taurus Orogenic Belt, the formation and geochemical features of Yeşilyurt and Kuluncak fluorites differ from each other. Yesilyurt fluorites are observed as vein type along the plane of unconformity between metamorphic rocks on hydrothermal phase. Kuluncak fluorites are observed in the contacts of alkaline syenites and marbles as vein type which the mineralization occured in the pegmatitic phase. Yeşilyurt fluorites are poor in terms of rare earth element contents but Kuluncak fluorites are rich. All Eu/Eu* values have negative Eu anomaly. This indicates that the fluorite created during the early phase is rich in terms of Eu^{+2} and that there is a presence of low temperature (T) and low oxygen fugacity ($_{f}O2$) in the environment.

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