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INVESTIGATIONS OF FISSIONABLE ELEMENTS IN THE TURKISH FLUORITES

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Fluorite is a calcium fluoride, CaF_2, a mineral. Although CaF_2 is a common mineral, fluorite is found in nature usually in small amounts in combination with some other minerals. These minerals can be classified as oxides, silicates, phosphates, carbonates and molybdates, etc. Fluorites contain rare-earth elements such as La, Ce, Nd, Sm, Y, etc., and other elements such as Mo, Zr, Ti, Fe, Sr, Ba, La, Ce, Nd, etc. Fluorite minerals of high purity seldom occur in nature. Except for the above mentioned elements fluorite is usually found in nature mixed with silicates and therefore they may contain Si, Al, Fe, Mg, Ca.

Fluorite is used for production of fluid slag in steel making, in smelting ore and as a constituent of white glasses and enamels. Fluorite is also used in cements, ceramics and plastics, glass industry and in making high octane fuels, freon and many other chemical products. Fluorite is a very attractive mineral of different colours and is often used as a gem. Since Turkey has world-biggest fluorite deposits, it is important to investigate fissionable elements (U and Th) content in all samples. In this investigation two methods of radiography have been used. One of them is neutron radiography for determination of the U and Th concentration using the irradiation of the samples by fast and thermal neutrons[1,2]. The other method is alpha autoradiography which was used for determination of the radioactive source type of the "stars", which represent several etched fission fragment tracks in Melinex film, emitted practically from one point, after the neutron irradiation of the fluorite sample (Fig.1). In this case polymerical films of different thickness for adsorbtion of alpha-particles with the maximum energy emitted from each studied isotope or their decay products were used [3].

EXPERIMENTAL TECHNIQUES

21 samples obtained from all fluorite deposits of Turkey were investigated. The number and the origin of the samples are presented in Table 1. Sample powders were put into Melinex plastic packets (3-7cm^2) for the neutrons exposition. For the neutron beam fluence determination thin natural ^{235}U and ^{238}U calibrated layers with the 0.1-0.4$\mu g/cm^2$ concentrations were used. Homogeneous calibrated thin layers of natural uranium and the investigated speciments have been irradiated by the fluence of $6x10^{12}$ thermal neutrons per $см^2$ at the MT-25 microtron of the FLNR JINR (Dubna). After the irradiation, fluorite samples were removed from the detector packets and all detectors were etched during 6 hours in 6.25N NaOH at 60°C. The enlarged fission fragment tracks were counted using an optical microscope, and the densities of induced fission tracks were determined.

After the neutron irradiation of the Melinex detector together with the sample N 10 (Table 1), the "stars" of fission fragment tracks have been observed (Fig. 1). For determination of the alpha- sources of these "stars" the radiographic analysis of the alpha-emitters using the CR-39 dielectric track detectors was carried out. Before the exposure the CR-39 detector (used for alpha-particles registration) was etched during ~

14 hours in 6.25N NaOH at 60°C to reveal background tracks, that are coming from the decay of radon from air, and for realiable identification of the investigated isotopes tracks. For registration of alpha particles emitted from natural U and Th, a thin sample of the fluorite sample N 10 was put in contact with the etched CR-39 during different exposition time: 1; 3; 5; 7 and 85 days. After the neutron exposition all detectors were etched during 6 hours in 6.25N, NaOH at 60°C. After detectors scanning under optical microscope the total number of the "stars" (Figure 2a) and the number of tracks in each observed "star" have been measured.

To determine the source type of the "stars" of the polyethylene terephtalate (PETP) films of a definite between the fluorite sample and detector was used to separate α particles emitted, from U and Th decay products. The decay schemes and the energies of the α particles emitted from ^{235}U, ^{238}U and ^{232}Th and their products are given below:

$^{235}U \rightarrow \alpha(4.6;4.4MeV) + ^{231}Th \rightarrow \beta + ^{231}Pa \rightarrow \alpha(5.0;4.7MeV) + ^{227}Ac \rightarrow \beta + ^{227}Th \rightarrow \alpha(5.0;4.7MeV) + ^{223}Ra \rightarrow \alpha(5.9;5.4MeV) + ^{223}Fr \rightarrow \beta + ^{219}Rn \rightarrow \alpha(6.8;6.56;6.43MeV) + ^{219}At \rightarrow \alpha(\underline{\mathbf{7.4MeV}}) + ^{215}Bi \rightarrow \beta + ^{215}Po \rightarrow \alpha(7.4MeV) + ^{211}Pb \rightarrow \beta + ^{211}Bi \rightarrow \alpha(6.6;6.3MeV) + ^{207}Tl \rightarrow \beta + ^{207}Pb$

$^{238}U \rightarrow \alpha(4.5;4.196MeV) + ^{234}Th \rightarrow \beta + ^{234}Pa \rightarrow \beta + ^{234}U \rightarrow \alpha(4.7MeV) + ^{230}Th \rightarrow \alpha(4.7;4.6MeV) + ^{226}Ra \rightarrow \alpha(4.8;4.6MeV) + ^{222}Rn \rightarrow \alpha(5.48MeV) + ^{218}Po \rightarrow \alpha(6.0MeV) + ^{214}Pb \rightarrow \beta + ^{214}Bi \rightarrow \alpha(5.5MeV) + ^{210}Tl ^{214}Bi \rightarrow \beta + ^{214}Po \rightarrow \alpha(\underline{\mathbf{7.8MeV}}) + ^{210}Pb$
$^{210}Tl \rightarrow \beta + ^{210}Pb \rightarrow \beta + ^{210}Bi \rightarrow \beta + ^{210}Po \rightarrow \alpha(5.3MeV) + ^{206}Pb$

$^{232}Th \rightarrow \alpha(8.0MeV) + ^{228}Ra \rightarrow \beta + ^{228}Ac \rightarrow \beta + ^{228}Th \rightarrow \alpha(5.4MeV) + ^{224}Ra \rightarrow \alpha(5.7MeV) + ^{220}Rn \rightarrow \alpha(6.3MeV) + ^{216}Po \rightarrow \alpha(6.8MeV) + ^{212}Pb \rightarrow \beta + ^{212}Bi \rightarrow \beta + ^{212}Po \rightarrow \alpha(\underline{\mathbf{8.9MeV}}) + ^{208}Pb$

The maximum energies of α-particles (E_{max}) emitted from ^{235}U, ^{238}U and ^{232}Th decay products and the range of these α-particles in PETP (R) are given below,

^{219}At (^{235}U) : E_{max} = 7.4 MeV, R=51.2μm
^{214}Po (^{238}U) : E_{max} = 7.8 MeV, R=54.4μm
^{212}Pb(^{232}Th) : E_{max} = 8.9 MeV, R=65.6μm.

For the separation of the ^{232}Th source an absorber with a thickness of ~54.5μ, located between the fluorite sample and detector was used. After a ~7 day exposition with a PETP absorber, the number of the "stars", their dimensions (radius of the "star" spot) and the number of tracks in each star have been measured using an optical microscope and the special automatic television system of the FLNR JINR [4].

RESULTS.

The fissionable elements concentration C in the fluorite samples has been calculated by the following equation

$$C^{238U}_{sample}(g/g) = \frac{C^{238U}_{calib.}\,\rho_{CaF_2}\,\varepsilon}{\rho_{calib}\,R_{eff}}, \qquad (1)$$

where ρ_{CaF_2} and ρ_{calib} is the density of tracks per cm^{-2} for the investigated and calibrated samples; respectively; ε is the etching efficiency which has a value of 80% for our etching conditions; R_{eff} is the effective range of fission fragments in CaF_2. The effective range for the complex material can be calculated by[5]

$$R_{eff} = 0.046 \sum_{i=1}^{n} a_i Z_i + 0.78, \qquad (2)$$

where a_i is the contamination of each element in the sample and z_i is the charge of atomic elements which the material contains. The calculated effective range of fission fragments for fluorite is 1.41mg/cm^2.

The concentration of fissionable elements in fluorite samples determined by (1) is,

$$C_{CaF_2}(g/g) = 1.123.10^{-7}\rho_{CaF_2}. \qquad (3)$$

The obtained results are presented in Table 1. The concentration of uranium elements is between ~10^{-5} and ~10^{-7} g/g and of thorium ones is between 10^{-4} and10^{-7} g/g. The minimum rate of Th/U is zero and the maximal value is 13. In the detector for the sample N10 large number of track "stars" was found. The U concentration in the "stars" region in the sample N10 is about 0.1-0.4g/g. In the sample N10 and 18 the Th element admixture is less 10^{-9} g/g.

To determine a source type of the observed track "stars" (Fig.1) after neutron irradiation, this sample together with a CR-39 detector has been exposed during different time. In the case of a 1 and a 3 day exposure, only a small number of "stars" with low track density was observed. For each exposure, the diameter and the number of tracks for each "star" was measured. For example, the distributions of the number of the "stars" according to the track quantity of each "star" and the radii of the "stars" for a 7 day exposition, are presented in Figs.3a and b. In the case of a 85 day exposure time a very high number of "stars" in the detector was found. All "stars" are located very close to each other and the number of tracks in each "star" and the diameter of the "stars" could not be measured accurately. In this detector about 254 stars/cm^2 have been observed. After a 7 day exposure without an absorber **66** "stars" in the area of 2 cm^2 were found.

TABLE 1. Results of investigations of the U-Th element concentrations in different fluorite samples by the neutron radiographic method.

Number	*Origin of The Sample*	C_{CaF2} (g/g)		C_{Th}/C_U
		U	Th	
1	Giresun-Sebinkarahisar	$7.7.10^{-6}$	$3.2.10^{-5}$	4.2
2	Giresun-Sebinkarahisar	$1.5.10^{-7}$	$6.0.10^{-7}$	4.0
3	Eskisehir-Beylikahir	$3.7.10^{-6}$	$2.0.10^{-5}$	5.4
4	Malatya-Kuluncak,Sofular Village	$9.3.10^{-6}$	$1.3.10^{-4}$	13
5	Giresun-Sebinkarahisar	$3.6.10^{-6}$	$5.8.10^{-6}$	1.6
6	Giresun-Sebinkarahisar	$8.6.10^{-7}$	$1.5.10^{-6}$	1.7
7	Giresun-Sebinkarahisar	$1.4.10^{-6}$	$2.4.10^{-6}$	1.7
8	Kayseri-Felahiye, Hayriye Village	$1.2.10^{-6}$	$8.0.10^{-6}$	6.6
9	Yozgat-Sefaatli, Cangili Village	$3.7.10^{-8}$	$1.4.10^{-7}$	3.8
10	Bursa-Uludag	$1.1.10^{-5}$	$2.6.10^{-7}$	≤0.03
11	Kirsehir, Payrenk Village	$2.0.10^{-7}$	$2.4.10^{-7}$	1.2
12	Kirsehir -Kaman, Alisar Village	$4.0.10^{-6}$	$7.0.10^{-6}$	1.8
13	Yozgat-Sefaatli-Garli Usagi Village	$2.0.10^{-7}$	$5.0.10^{-7}$	2.5
14	Yozgat,Hacili Village	$1.5.10^{-7}$	$8.5.10^{-7}$	5.7
15	Yozgat- Sargun-Emirhan Village	$2.0.10^{-7}$	$6.0.10^{-7}$	3.0
16	Kirsehir, Karaman-Yeniyapan Village	$1.7.10^{-7}$	$8.3.10^{-7}$	4.9
17	Kayseri-Felahiye	$4.6.10^{-5}$	$2.7.10^{-4}$	5.9
18	Kirsehir -Akcakent, Istanbullu	$2.0.10^{-7}$	~ 0	
19	Yozgat, Akdag Madeni-Y. Culha Village	$5.4.10^{-6}$	$7.6.10^{-6}$	1.4
20	Kirsehir, Kaman-Durmuslu Village	$1.8.10^{-7}$	$1.6.10^{-7}$	1.0
21	Yozgat, Bogazliyan, Yavasli Village	$8.2.10^{-6}$	$1.6.10^{-5}$	2.0

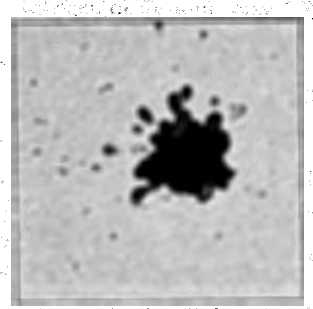
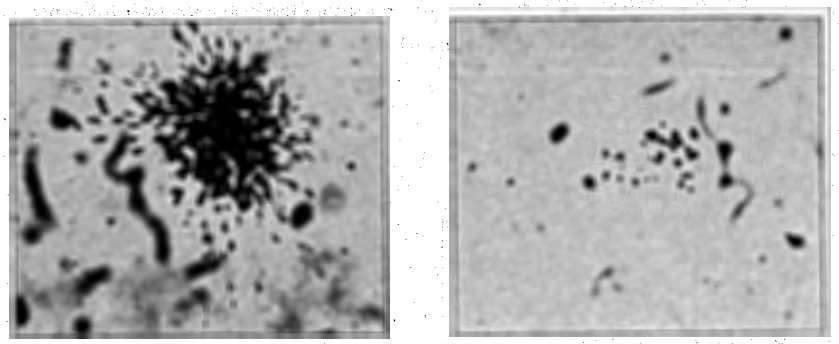


Figure 1. Microphotograph of the "stars" of the etched fission fragment traks in the Melinex detector after neutron irradiation of sample N10.



a b

Figure 2. Microphotographs of the "stars" of the etched alpha particle tracks in the CR-39 detector for a 7 day exposition without an absorber(a), and with 54.4μ PETR absorber(b).

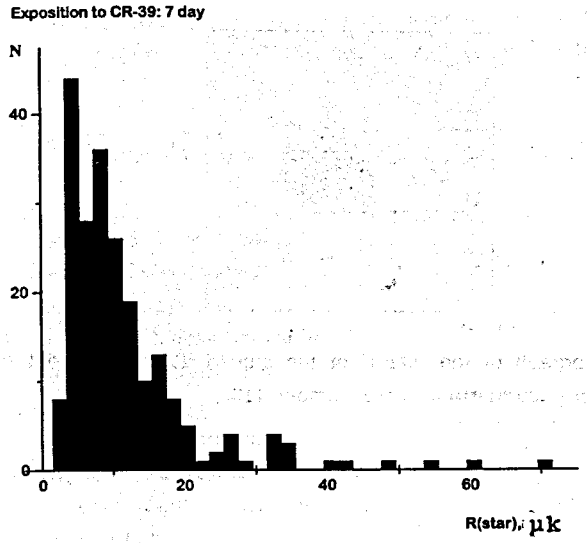


Figure 3a.Distribution of the"stars" number according to the radius R of each "star".

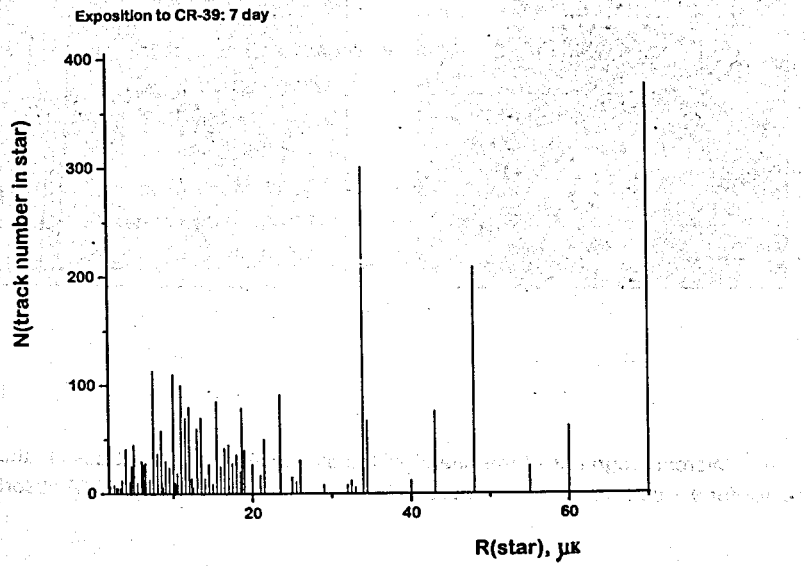


Figure 3b.Distribution of the etched alpha particle tracks number in each "stars" with radius R, after 7 day exposition to the CR-39.

Using a PETP absorber with a thickness of 54.4μm only 1 "star" in the same area was observed (Figure 2).

CONCLUSIONS

As a result of the performed investigation of the fissionable element concentrations in the examined fluoride samples by neutron radiography the obtained U-Th contents vary in the range of10^{-4} - 10^{-7} g/g. The Th/U ratio for 8 samples did not exceed 2, for 9 samples ≤7 and only for sample N4 this value was 13. For the irradiation conditions of this experiment, the sensitivity of the used methods is 10^{-9}g /g. The results of α-autoradiography show that with this method it is possible to separate Th and U elements using the CR-39 detector.

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REFERENCES

1. Fleischer R.L., P.B. Price and R.M. Walker, In: Nuclear Tracks In Solids: Principles and Applications, (University of Califonia Press, Berkeley, 1975).

2 Flerov G.N., Bersina I.G. In: Radiograpghy of minerals, rocks and ores.(In Russian). M.: Atomizdat,1979, p.123-126.

3. Victorova N.N. et al, Nucl.Tracks Radiat.Meas., 1993, v.22, N1-4, pp.885-888.

4. Ponomarenko V.A. et al , Radiat.Meas., 1995, v.25, N1-4, pp.769-770.

5. Abdullaev X. et al., Preprint JINR, P 13-3243, Dubna, 1967.

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