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$K_2Ca_2(SO_4)_3$: Eu POWDER FOR ENVIRONMENTAL RADIATION MONITORING WITH TL ULTRASENSITIVITY POWDER

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An original procedure to obtainia the high purity $K_2Ca_2(SO_4)_3$:Eu power is presented: the optimal choice of reagents and their ratios, the chemical procedure as well as the thermal and mechanical treatment procedures are detailed. The final characteristics of this powder are: nonhigroscopic, stable white crystals. The preliminary controls and the obtained results are as follows: the glow curve shape obtained in a 662 keV gamma ray irradiation field shows the maxim glow peaks at temperatures 70, 90, 130 degrees Celsius; the range of measurement of the absorbed dose is $(0.5x10^{-6}$- 10) Gy. The determinations were carried out by a TLD reading system type. The whole dosimetric system was calibrated by using a ^{90}(Sr+Y) standard source. The fading values of information are about 3 % in a 30 days interval.

1. Introduction

The absorbed dose in air and its rate are the most used dosimetric quantities , as they refer to the ermetical transfer from the ionizing radiations to the environment.

Some of the most used methods for their measurement rely on the thermoluminiscent dosimetric systems due to their special advantages as follows: the dosimetric information can be integrated on the whole exposition period, the detectors can be reused for a large number of cycles; easy to use, the loss of information during long periods, (until 60 days is negligible); the detectors are very sensitive their response being constant along very wide rances of doses.

In our laboratory there is an important experience in obtaining and use the TL detectors based on LiF:Mg,Ti (natural Li); CaF_2:Mg; CaF_2:Mn; $CaSO_4$:Dy. The $CaSO_4$ detectors are normally used for monitoring the environmental absorbed doses within the range $(3.10^{-5}$ - 10)Gy [1,2].

As the new standardization documents require limits of measurements the lowering of the inferior new dosimetric systems, having superior sensitivity is studied [3].

One of these new thermoluminophors is $K_2Ca_2(SO_4)_3$:Eu thermoluminophore, as powder, and the results obtained in its characterizations.

Its sensitivity is 2-4 times superior to the $CaSO_4$:Dy powder [4]; the glow curve presents 3 peaks. The measurement interval was extended to $(0.5\cdot10^{-5}$-10)Gy and the measurement errors were below the limits imposed by the ICE 1066/91 [5].

The most important disadvantage of this luminophore is due to its high effective atomic number value, Zeff = 11.3 compared to air Zeff = 7.2, and as a consequence a strong energy dependence of the response. This dependence can be attenuated be the use of metallic filters [6].

2. Thermoluminophore preparation

The chemicals used for the preparation of the powder were: ultrapure K_2SO_4 and $CaSO_4 \cdot 2H_2O$ and $EuCl_3$ mody by Fluka.

The water used for dissolving was deionized, triple distilled on quartz glassware. Quartz and platinum oven were used, and two separate solutions were obtained.

One of them was prepared by dissolving K_2SO_4 in tridistilled water in stoechiometric ratio while the second was obtained by dissolving $EuCl_3$ in a small quantity of tridistilled water.

The mixture solution, obtained from the two solutions, was boiled in a quartz vessel continuously stirred, while the whole quantity of water evaporated.

A crystalline, brilliant powder of K_2SO_4:Eu was obtained. It was crushed in a having agat balls and mixed with dehidratated $CaSO_4$ in a 1:2 ratio by weight and then homogenized with a teflon sticke..

The mixture powder was then calcinated at 980^{o}C for a 26 hour period in a platinum crucible with rapid heating and cooling at the ambient temperature in order to obtain a stable phosphor mesh. Crystals having a diameter ϕ =100µm were used for subsequent studies.

3. Results and discussions

Comparative studies regarding the main characteristics of both thermoluminophores $CaSO_4$:Dy and $K_2Ca_2(SO_4)_3$:Eu were accomplished.

The first studies regarded the comparative glow curves shapes and the possibilities to eliminate the secondary peaks.

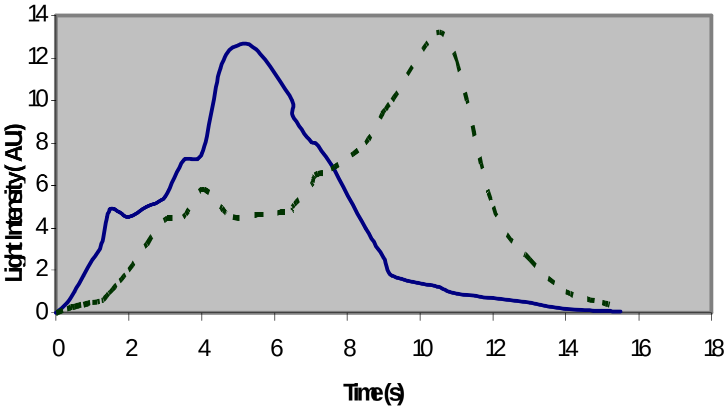

Fig.1. Glow curve analysis (___ $Ca_2K_2(SO_4)_3$:Eu; ----- $CaSO_4$:Dy)

The thermoluminophore powder was irradiated with absorbed dose values of 0.3 Gy and 0.6 Gy generated by a 1.4 GBq, ^{90}(Sr+Y) source.

Reading of the information was made by a TLD reader MARK IV type, Model 1100, provided with an X-Y recorder. The reader has a linear heating cycle up to 400°C with a heating rate of 15°C/s. Fig 1 presents the shapes of the two glow curves.

$CaSO_4$:D shows two peaks: the main peak at 220°C and the secondary one at 100°C.

$K_2Ca_2(SO_4)_3$:Eu has a main peak at 130°C and two secondary peaks at 70°C and 90°C respectively The presence of these secondary peaks is due to some surface luminophore centers.

The secondary peaks were removed followed by using several cycles: irradiation with an absorbed dose of about 0.1 Gy followed by thermal treatment (regeneration) at 330°C temperatures for 1 hour period slow cooling. These peaks were totally removed in the case of $K_2Ca_2(SO_4)_3$:Eu luminophores, while in the case of $CaSO_4$:Dy the removal was partial.

The dosimetric characteristics were checked by measuring the response of the TLD system within a large range of absorbed doses. These reference values of dose were obtained with two radioactive sources: a ^{137}Cs source with 140.6 MBq activity and any^{60}Co, with the 81.4 GBq activity.

The irradiation was performed in a panoramic irradiation geometry. The measurement errors, ε [%], were calculated according to the relation: ε [%]=(MD-CTD)/CTD, where MD id the measured dose and CTD is the conventionally true doses respectively.

Table 1
The obtained results:

CTD Gy	$CaSO_4$:Dy			$K_2Ca_2(SO_4)_3$:Eu		
	E[imp]	MD[Gy]	ε [%]	E[imp]	MD[Gy]	ε [%]
0.5×10^{-5}				30	0.42×10^{-5}	-15.1
1.0×10^{-5}	38	1.52×10^{-5}	+52.0	82	1.16×10^{-5}	+15.8
3.0×10^{-5}	58	2.30×10^{-5}	- 22.0			
5.0×10^{-5}	100	4.00×10^{-5}	-20.0	280	3.95×10^{-5}	-21.0
1.0×10^{-4}	225	0.90×10^{-4}	-10.0	650	0.918×10^{-4}	-8.0
1.0×10^{-3}	3000	1.20×10^{-3}	+20.0	7500	1.05×10^{-3}	+5.0
1.96×10^{-2}	5100	2.04×10^{-2}	+4.0	17730	2.50×10^{-2}	+21.6
1.0×10^{-1}	27000	1.08×10^{-1}	+8.0	63000	0.89×10^{-1}	-11.0
2.2×10^{-1}	54000	2.00×10^{-1}	-9.0	130000	1.84×10^{-1}	-16.0

Another check was made in order to evaluate the upper fading limit the $K_2Ca_2(SO_4)_3$:Eu during a 30 days period.

Six different thermoluminophore samples, containing each 0.065g of powder were irradiated with an absorbed dose of 10^{-2} Gy using a ^{137}Cs source with 100 GBq activity .

Three of the samples were read at one hour after the irradiation and the other three were read after a 30 day period. The obtained dose values were the following:
(MD) 1 hour = 1.003×10^{-2} Gy (710 imp/Gy);
(MD) 30 days= 0.991×10^{-2} Gy (702 imp/Gy).
The difference represents the loss of information in the limit of 1.2 % , which is a very satisfactory result.

Conclusions

Based on the above,the following conclusions are evident: • an original procedure, based on the use of ultrapure chemicals was developed, in order to obtain a high quality $K_2Ca_2(SO_4)_3$:Eu thermoluminophore; • the new luminophore is very sensitiv 2-6 times higher than $CaSO_4$:Dy luminophore; • the very low fading effects allow its exhibit in environment for long period measurements.

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