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ABSTRACT

Three common thermoluminescent phosphors, $CaSO_4$:Dy, CaF_2:Dy, and CaF_2:natural, have been examined to determine the feasibility of their application in thermal neutron dosimetry. Basically, the phosphor is exposed to thermal neutrons (and other radiations) causing activation of nuclei in the phosphor. Next, the phosphor is stored to undergo self-irradiation from the internal radioactive nuclei, in this case all beta emitters. Later one reads the thermoluminescence induced by the beta emission during storage. This reading depends on the original neutron exposure, and could thus provide a dosimetry system. The phosphors contain suitable isotopes ^{44}Ca or ^{164}Dy (and ^{34}S). Limits for the lowest detectable fluence were determined by extrapolating results from high fluence measurements. Using the decay of ^{165}Dy (2.3 h half-life) the lowest detectable fluence is estimated to be 5×10^7 n/cm^2. Using ^{45}Ca decay (165 d half-life), this limit is raised to about 5×10^{10} n/cm^2. Methods for improvement are discussed.

INTRODUCTION

Detecting thermal neutrons in mixed radiation fields necessarily relies on the neutron's nuclear interaction with the detector. The useable nuclear interactions might be roughly divided between those which happen rapidly, therefore leaving their mark during irradiation, and those of simple activation wherein the product is detected after irradiation. An example of the rapid type is the $^6LiF/^7LiF$ thermoluminescent system which detects[1] the neutron by the energy deposited from the (n, α) reaction in ^6Li. The ^7LiF is used to measure and correct for the energy deposited by the other radiations. Metal foils provide an example of detection by simple activation. In this case the neutrons produce radioactive nuclei in the foil, and these are later detected upon their disintegration, usually by beta counting. This technique requires no correction for effects of other radiations.

A thermoluminescent system based on simple activation, like foils, has been studied earlier at this laboratory.[2,3] Natural CaF_2 was exposed to thermal neutrons in a mixed field, the neutrons causing activation of ^{44}Ca to ^{45}Ca, and the other radiations inducing thermoluminescence (TL). Next, the samples were annealed to eliminate the TL induced during irradiation, and then they were stored to undergo self-irradiation as betas are emitted from the radioactive ^{45}Ca. Finally, the TL was read after a suitable storage time. This final TL reading is related to the number of disintegrations occuring during storage which in turn depends on the original neutron fluence. Like foils, the signal is independent of the other radiations in the mixed field provided that the crystal does not suffer permanent radiation damage. An advantage over foils is that integration of the decay betas occurs outside the detecting apparatus.

Herein we further examine the feasibility of using phosphor activation as a thermal neutron dosimeter. Our purpose is to establish the lower limits for the detectable fluence with CaF_2:natural, and to extend the study to $CaSO_4$:Dy and CaF_2:Dy. The ^{164}Dy in the latter two phosphors might provide a thermal neutron dosimeter useful in accident situations.

EXPERIMENTAL

The CaF_2 was collected from a mine located in Santa Catarina State, Brazil and was ground and seived through 80 onto 200 mesh Tyler screens for use. This powder can be very

sensitive to light so that a 10 second exposure to incandescent room light could be detected. Since work under darkroom conditions was not convenient, the samples were annealed in air for 45 min at 570°C whereupon the light induced TL was just detectable after a 10 min exposure to room light. Sensitivity to gamma-rays is also reduced by the annealing treatment. Several batches of about 2 grams each were prepared and two were selected for further study on the basis of their similar response to gammas. Light was normally avoided except for subdued room light during the reading process.

The CaF_2:Dy (TLD-200) and $CaSO_4$:Dy samples were purchased from Harshaw Chemical, the former stated to be 80 to 200 Tyler, and the latter found to pass 80 mesh and not 200 mesh. The CaF_2:Dy was annealed at the highest reading temperature before use, nominally 400°C. The $CaSO_4$:Dy powder was initially annealed 90 min at 700°C, then at 400°C for reuse.[4]

Gamma irradiation was at two meters from a 40 Ci ^{137}Cs source, and mixed field irradiations occured at a pneumatically served station alongside the core of IEAR-1, the swimming pool reactor located at this Institute. The neutron flux at this station is about 2×10^{12} n/cm^2 -sec as determined by gold foil activation measurements. Samples were exposed in cylindrical polyethylene capsules having a 1 mm wall and 3 mm inside diameter.

Glow curves were recorded as photocurrent against time on two Harshaw model 2000 systems, both with the phototubes at room temperature. The relative sensitivity of the two machines was compared by reading identically treated LiF dosimeters in both instruments. All measurements in this work were made by dispensing a fixed volume of powder. The corresponding masses are CaF_2:Dy, 20.1 mg; $CaSO_4$Dy, 18.9 mg; and CaF_2:natural, 20.7 mg, with the total fluctuation at about 0.3 mg. Although the Harshaw readers do heat the sensing thermocouple linearly, a particular peak's position is not reproducible unless the reading cycle is rigidly controlled: time of initiation, time of termination, and time of drawer open and close. Since these conditions were not met for our measurements, relative peak positions between different glow curves are only approximate. The heating pan's temperature was determined for a typical cycle by spotwelding a thermocouple to an old pan and registering the temperature against time. This temperature curve is not strictly linear, as reflected by the slight nonlinearity in the temperature scale of Fig. 1.

RESULTS AND DISCUSSION

a) Gamma-Ray Irradiation

The TL induced by a one R gamma exposure is shown in Fig. 1 for equal volumes of CaF_2:Dy (dot-dashed line), $CaSO_4$Dy (dashed line), and CaF_2:natural (solid line). The shapes of the glow curves are similar to those observed by others.[4,5,6] For CaF_2:Dy and $CaSO_4$:Dy the height of the highest peak was used as a measure of the TL. For convenience, the three peaks in CaF_2:natural were labled 1, 2, and 3 in ascending temperature order, and normally changes in both peaks 2 and 3 were followed, again using the peak height. On this basis CaF_2:Dy gives about 30% more TL than the same volume of $CaSO_4$:Dy, as shown in Fig. 1. The response of CaF_2:natural is about ten times smaller; hence, its glow curve has been increased a factor of ten in the figure. The response of CaF_2:natural might be increased if the high temperature annealing were in N_2 and not air.[7]

b) Beta-Ray Irradiation from Internal ^{45}Ca (and ^{35}S)

When calcium comporids are exposed to thermal neutrons, some of the ^{44}Ca (σ = 1.1b, natural abundance = 2.06%) is activated to ^{45}Ca a beta emitter with a 165 d half-life. To observe our phosphors' response to this internal activity, we irradiated a sample of each for 1 min in IEAR-1. Several days later, phosphor was placed in the reading pan and heated one

minute at the maximum temperature, nominally 400°C. (The photo tube was inactivated during this heating, otherwise the high ligh levels spoil the subsequent low level readings). The samples could then be read at various intervals to determine the response as a function of the self-irradiation time. This technique has the advantage that spurious luminescence is largely eliminated since the powder is not handled between readings.

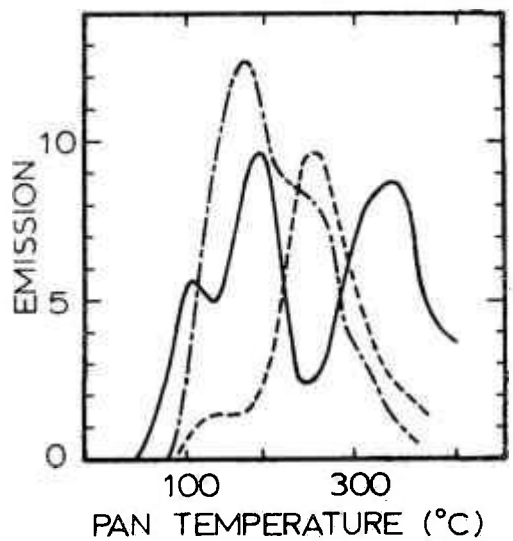

Fig. 1
The TL induced by a 1 R exposure to ^{137}Cs gamma. Dot-dashed line, CaF_2:Dy. Dashed line, $CaSO_4$:Dy. Solid line, CaF_2:natural.

The responses of the three phosphors are shown as a function of the self irradiation time in Fig. 2. In contrast to gamma irradiation, $CaSO_4$:Dy gives about 5 times more TL than either CaF_2:Dy or CaF_2:natural (peak 3). The shapes of the glow curves are largely unchanged from those shown in Fig. 1, although peak 2 in CaF_2:natural is relatively smaller compared with peak 5, as indicated in Fig. 2. Some of the response in $CaSO_4$:Dy can be attributed to ^{35}S (88 d half-life) created during irradiation from ^{34}S (σ = 200 mb, natural abundance = 4.22%). Assuming that the TL is proportional to the total decay energy times the number of disintegrations, one finds that the ^{35}S contribution in $CaSO_4$:Dy would be about 70% that of ^{45}Ca during the first 10 days. Therefore, considering only the ^{45}Ca decay, $CaSO_4$:Dy is about 3 times more sensitive thant the other two phosphors. (We suppose throughout that interactions with fast neutrons are negligible.) The greatly increased relative response of CaF_2:natural may correlate with an increased sensitivity to light. (See below.) The linear response is expected in all cases since the times are short compared with the half-lives.

Any application of these materials to high fluence dosimetry requires knowing the response as a function of irradiation time (i.e. fluence). The linear response in Fig. 2 leads us to expect a linear behavior with number of disintegrations and hence with neutron exposure as well. Since samples exposed to high fluence cannot be reused, verifications of the response

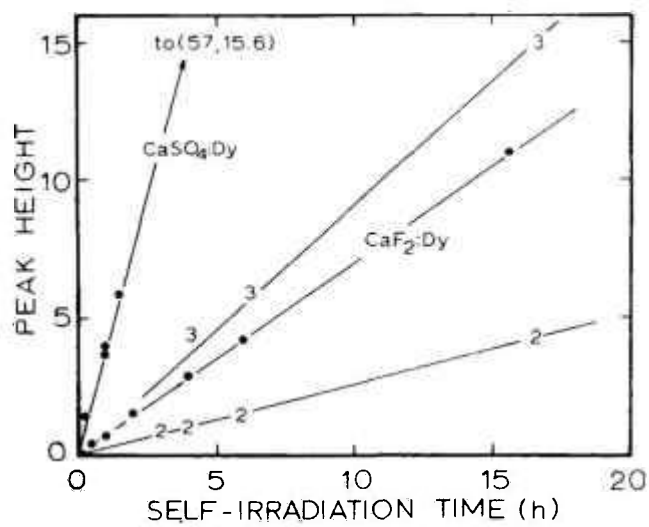

Fig. 2
The growth of the TL as a function of the self-irradiation time, for a 1 minute irradiation.

concentrated on CaF_2:natural because it is inexpensive. In a typical experiment, samples of CaF_2:natural were irradiated for 0.5, 1, 2, 4, and 6 min in IEAR-1, and then after several days they were annealed at 400°C, stored in capsules, later read, annealed again, stored for a different time, read, and so forth. In Fig. 3 the heights of peak 2 and 3 are shown as a function of the original irradiation time, for readings taken after 16 h storage. The response is linear for peak 2, as expected, and peak 3 also appears to respond linearly though there is more fluctuation. The high exposure inherent in the irradiation increases the light sensitivity of CaF_2:natural the order of 10 times. This increase does not depend strongly on irradiation time 0.5 to 6 min, and probably corresponds to the filling of deep traps originally emptied by annealing. Filling such deep traps could cause the increased relative sensitivity of CaF_2:natural to internal betas.[6]

In any case, the increased light sensitivity cannot account for the poor reproducibility generally encountered in these experiments. (Figure 3 is the best run.) Typically two out of five capsules, either with ascending or identical irradiations, would display unsuitable response despite similar handling of all samples. Sometimes peak 3 would be at about the expected level, but peak 2 much higher than expected, and sometimes both peaks would be higher than expected. Similar problems were not encountered with CaF_2:Dy and $CaSO_4$:Dy, but these were not used extensively.

Despite these problems, we can establish a limit for the lowest detectable fluence by making the linear extrapolations implied in Figs. 2 and 3. For $CaSO_4$:Dy the signal after 0.5 h and 1 min irradiation (Fig. 2) is about 9 times larger than the smallest detectable one. Since the fluence in this case is about 1.2×10^{14} n/cm^2 we conclude that for 0.5 h storage the lowest detectable fluence would be about 1.3×10^{13} n/cm^2. To detect lower fluences the storage time must be increased, although this recourse also has limits. For these long half-life decays the fundamental limit is that the internal activity must induce TL at a rate comparable to that due

to back-ground radiation and natural internal activity. Typically, this background is equivalent to the order of 0.5 mR per day, and we rather arbitrarily define the detection limit as 20% above the background, or the equivalent of 0.1 mR per day. The growth observed for $CaSO_4$:Dy in Fig. 2 is the gamma equivalent of about 6.8 mR/h. Reducing this rate to 0.1 mR per day implies a reduction of the detectable fluence to about 4×10^{10} n/cm^2 (40 rem). For CaF_2:Dy the value would be about 2.5×10^{11} n/cm^2 (5 times less TL per neutron, 30% higher sensitivity to back-ground). These estimates are only valid if the sensitivity to back-ground remains unchanged after the neutron exposure. Since the relative sensitivities change this may not be the case. The CaF_2:natural can be supposed to change sensitivity, as evidenced by changing light response.

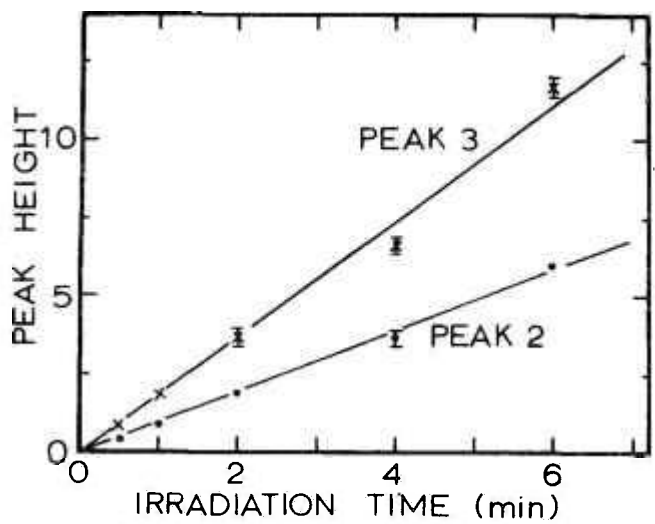

Fig. 3
The TL as a function of the original irradiation time for a 16 h storage time. (This data is not typical - see text).

In any case the lowest detectable fluence of 4×10^{10} n/cm^2 (about 40 rem) for $CaSO_4$:Dy can be considered as an order of magnitude boundary for use of Ca (and ^{34}S) activation as a dosimeter. Isotope enrichment could provide an improvement by a factor of about 45, and storage in low background environments could also improve sensitivity. Employing this system at high fluence may be useful since other TL systems saturate. Exposing TLD-100, 600, and 700 1 min in IEAR-1 gives glow curves clearly in the saturation region, although the curve's shape might still give the absorbed dose.[8] To reach the low fluence region useful for personnel dosimetry, a method must be found to obtain more TL per disintegration, or the system must rely on a different isotope.

c) Beta-Ray Irradiation from Internal ^{165}Dy

Like ^{44}Ca, ^{164}Dy (σ=2600b, natural abundance = 28.2%) can produce TL after activation to ^{165}Dy, the latter a beta emitter with a 2.32 h half-life. To observe TL due to decay of this isotope, a recently irradiated sample was placed on the reading pan and read every

half-hour, without moving the sample. Figure 4 shows the decay in the induced TL as a function of time elapsed since a 1 min irradiation in IEAR-1. Each point for CaF_2:Dy is multiplied by 10 to eliminate overlap of the two curves. The decay clearly follows the 2.32 h half-life for ^{165}Dy which is indicated by the solid lines. In the final stages both samples leave the exponential decay as longer half-life isotopes begin to influence the response. For $CaSO_4$:Dy the experimental points are initially slightly above the ^{165}Dy decay line, indicating that another short half-life impurity may be present. Extrapolating to the irradiation time, and comparing with the ^{45}Ca results shown in Fig. 2, we can conclude that in the first 0.5 h the ^{165}Dy decay produces at least 7 x 10^3 more TL than ^{45}Ca. In one day the ^{165}Dy decay would produce approximately 1 x 10^3 more TL than ^{45}Ca. This would lower the limit for the detectable fluence to about 4 x 10^7 n/cm^2 (40 mrem) for either $CaSO_4$:Dy or CaF_2:Dy (30% higher), again supposing that the sensitivity to background radiation is not changed. Also, we have supposed that there is no time delay between irradiation and the start of storage. In practice this could only be realized by irradiating at high temperature to eliminate the intermediate anneal. Finally, since this case is not just a comparison of background and self-irradiation rates, the phosphor must be sensitive enough to detect the 0.1 mR supposed as the daily detection limit.

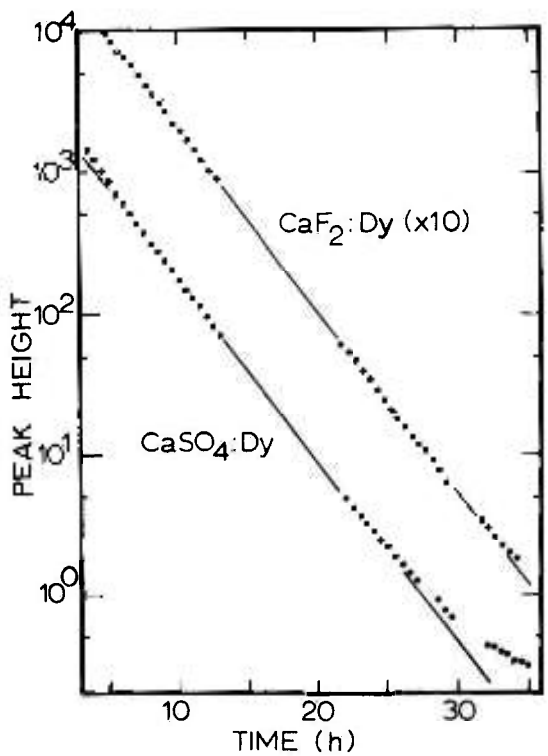

Fig. 4
The TL induced in a 0.5 h self-irradiation as a function of time since the original irradiation. The solid lines represent decay with a 2.32 h half-life.

Several improvements on the ^{164}Dy system are possible. A factor of about 3 could be obtained by isotope enrichment. More important, the Dy concentration might be increased, or the system might be changed to a mixture of some Dy compound and a TL phosphor. Significant interaction would be expected between the two powders since the maximum beta range is about 2.1 mm (in $CaSO_4$).[9] (Similar interaction from ^{45}Ca betas is small, as we have observed, because the maximum range in $CaSO_4$ is about 0.15 mm.) A mixture of Dy_2O_3 and LiF, for example, would have the advantage of being reuseable, even after exposure to high fluence.

RESUMO

Foram examinados três fósforos termoluminescentes comuns, $CaSO_4$:Dy, CaF_2:Dy e CaF_2: natural, afim de determinar as suas aplicabilidades na dosimetria de nêutrons térmicos. As amostras são expostas a nêutrons térmicos (e outras radiações), que ativam alguns dos núcleos desses fósforos. Em seguida, as mesmas são armazenadas, havendo auto-indução de termoluminescência devido ao fato das amostras serem irradiadas com raios beta dos núcleos ativados. A termoluminescência é então medida e, como o seu valor está relacionada com a exposição inicial de nêutrons, pode ser utilizada para dosimetria. Os fósforos contém núcleos aproveitáveis de ^{44}Ca ou ^{164}Dy (e ^{34}S). Os limites de fluência detetável foram determinados extrapolando-se os resultados em fluências mais altas. Considerando o decaimento de ^{165}Dy, com meia vida de 2,3 horas, calcula-se que a fluência mínima detetável seja da ordem de 5×10^7 n/cm^2. No caso do decaimento de ^{45}Ca de meia vida 165 dias, êste limite aumenta para aproximadamente 5×10^{10} n/cm^2. Possíveis melhoramentos são discutidos.

RÉSUMÉ

Trois phosphors thermoluminescents communs, $CaSO_4$:Dy, CaF_2:Dy et CaF_2:naturel ont été étudiés afin de déterminer la possibilité de leur application en dosimetrie de neutrons thermiques. Tout d'abord, le phosphore est exposé aux neutrons thermiques (et à d'autres radiations) provoquant l'activation des noyaux dans le phosphere. En suite, le phosphore est stocké pour subir une self-irradiation aux noyaux internes radioactifs, dans ce cas, émetteurs beta. La thermoluminescence induite par l'émission beta pendant le stockage depends de l'exposition initialle des neutrons, et peut ainsi tournir un système de dosimetrie. Les phosphores contiennent les isotopes utilisables ^{44}Ca ou ^{164}Dy (et ^{34}S). Les limites pour le plus faible flux dectable furent déterminés par extrapolation des résultats des mesures de haute flux. En utilisant la décroissance de ^{165}Dy avec le periode de 2,3 heures, le plus faible flux detectable est estimé à environ 5×10^7 n/cm^2. En utilisant la décroissance du ^{45}Ca de periode 165 jours, cette limite est d'environ 5×10^{10} n/cm^2. Des méthodes d'amelioration sont discutées.

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