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ABSTRACT

The technique of activation of thermoluminescent dosimeters can be used to detect any radiation which leaves the dosimeter radioactive. In this experiment $CaSO_4$:Dy purchased from Harshaw Chemical Co. was irradiated in the reactor of the Institute of Atomic Energy. When the effect of thermal neutrons was to be avoided, as in the case of fast neutron dosimetry, the dosimeter was wrapped in Cd. The initial step in the activation of the phosphor is the production of radioactive ^{32}P by the reaction $^{32}S(n,p)^{32}P$, which has an effective cross-section of 265 mb in the region of 2.7 Mev. The subsequent decay of the ^{32}P (with half life of 343 h) induces TL in the $CaSO_4$:Dy. The TL induced during the irradiation, caused principally by gamma-rays, is eliminated by annealing at a suficiently high temperature (600°C for 30 minutes), immediately following the irradiation. The self-induced TL resulting from the ^{32}P decay is a measure of the fast neutron fluence to which the dosimeter was exposed. The estimated lowest detectable fluence of neutrons with energies in the region of the reaction ($E \gtrsim 2.7$ MeV) is equivalent to one rad of Kerma in tissue.

Activation of thermoluminescent (TL) phosphors has been shown to be a potentially useful method for measuring thermal neutron doses, especially where mixed radiation fields and large scale distribution are important (MAYHUGH et al.). This technique consists of exposing a TL phosphor to thermal neutrons which activate nuclei in the material. After exposure the dosimeter is annealed to eliminate any TL induced during irradiation, and then stored to undergo self-irradiation from the internal radioactivity. A final reading gives the TL resulting from self-irradiation, and this measurement is directly related to the original thermal neutron fluence.

Obviously, the activation technique can be used to detect any radiation which leaves the dosimeter radioactive. Some threshold reactions for fast neutrons are an example of this extension, as others (PEARSON et al.) are also noting. The advantages for fast neutron detection are essentially the same as those for thermal neutron dosimetry; first, the dosimeter may be used in practically any mixture of radiations, provided it does not suffer damage; and, second, integration occurs outside the reading apparatus, in contrast to direct detection of radioactive decay as in beta-counting. Energy dependence necessarily follows the energy variation of the reaction cross-section.

To illustrate the feasibility of this scheme in mixed fields, we irradiated $CaSO_4$:Dy (Harshaw) in the swimming-pool reactor at this Institute. The reactor operates at 2MW and produces fast fission neutrons plus the order of 10^8 R/h of photons and about 10^{12} thermal neutrons/cm^2/sec at our irradiation station. When fast neutrons were to be detected, the dosimeter was wrapped in 1mm of Cd to avoid activation due to thermals.

Our results demonstrate that the activation technique is useful for fast neutrons even in this extreme mixed environment. Soon after a ten minute irradiation, a portion of the exposed powder was placed in the reading pan of a TL detector (Harshaw Model 2000) and was annealed by heating to 400°C and holding this temperature for one minute, all with the phototube deactivated. The TL induced by self-irradiation was then read daily for several weeks without disturbing the sample, and therefore without altering the sample's geometry. Until a subsequent 600°C anneal (see below) the measurements were complicated by a constant phosphorescence which was subtracted to give the TL due to self-irradiation.

The daily TL readings are summarized in Fig. 1, and they are seen to decrease at a rate characteristic of the 343 h half-life of ^{32}P which occurs through the threshold reaction $^{32}S(n,p)^{32}P$. The discontinuity at about 225 h results from removing the sample and annealing it for 30 min at 600°C. This procedure eliminates the troublesome phosphorescence, but apparently reduces the phosphor's sensitivity. The data are presented without normalization at this discontinuity because it illustrates the importance of a standardized anneal. In fact, the TL readings were not always taken at exactly twenty-four hour intervals, and the points shown in the main part of the figure were adjusted by dividing the response by $\exp(\Delta t/\tau)$-1 where Δt is the time in hours since the previous thermal emptying and τ is the mean lifetime for ^{32}P. Simply plotting the TL/Δt gives similiar results, but with more scatter. We conclude that from the third to at least the 30th day, the TL is caused by beta emission from ^{32}P induced by fast neutrons ($E \gtrsim 2.7$MeV).

In the hours just after irradiation, the TL response is dominated by decay of ^{165}Dy, as shown in the insert to Fig. 1. (Here the TL is recorded after 30 min self-irradiations without any normalization). Activation of ^{164}Dy occurs because its absorption cross-section is appreciable at intermediate energies where the Cd wrapping is no longer opaque to the incident neutrons. If only response to ^{32}P is to be integrated, the results show that the initial anneal should occur about two days after the irradiation so as to avoid contributions from shorter half-life species.

The fast neutron spectrum at the irradiation station was not accurately measured, and we cannot extrapolate the fast neutron response to lower fluences; hence, the experiments do not determine the lowest detectable fluence. This is not important, however, since the lowest detectable fluence is easily estimated.

For nuclear decays of relatively long hal-life (several days or more), the lowest detectable fluence depends on the relative rates of self-irradiation and irradiation from background sources. Background can easily be reduced to about 50μR per day (JONES et al.) which is presumably due to high energy cosmic rays and therefore corresponds to about 50μrad per day in $CaSO_4$. If self-irradiation produces 0.1mrad per day initially, then after one ^{32}P half-life (~ 14d), background will have contributed 0.7mrad, and self- irradiation 1 mrad. Measurement at these levels should not be very difficult with $CaSO_4$:Dy because of its high sensitivity.

A rate of 0.1 mrad per day means that 6.25×10^3 MeV must be deposited per gram per day. The average desintegration of ^{32}P will supply about 0.8 MeV, half the maximum beta energy of 1.71 MeV; hence, we require about 7.8×10^3 desintegrations per day per gram. (Here we assume that the sample can be made large enough to absorb essentially all the betas.) In the first day this energy will be supplied if 1.6×10^5 ^{32}P nuclei are present per gram. If the effective cross-section for activation of ^{32}S is taken as 265mb, the cross-section per gram of $CaSO_4$ is 1.1×10^{-3} cm^2/gm. This gives one ^{32}P nucleus per gram for about every 900 neutrons incident/cm^2. On this basis, obtaining 0.1 mrad in the first day of self-irradiation requires an original fluence of about 1.5×10^8 neutrons/cm^2. If lower rates of self-irradiation could be distinguished from background, a corresponding reduction in the lowest detectable fluence would result. In the reaction region (E $\gtrsim$ 2.7MeV) our estimate of the lowest detectable fluence corresponds to about 1 rad of kerma in tissue. For activation by thermal neutrons, similar estimates agree with the extrapolated high fluence values (MAYHUGH et al.) to within a factor of 2.

The above rate considerations indicate that given a certain concentration of radioactive nuclei, short half-life (a few hours) systems would be more sensitive in terms of the lowest detectable fluence, even though longer self-irradiations can be used in long half-life cases. for short half-lives the phosphor may not be so sensitive as to see the background during the shorter self-irradiation in which case the sensitivity would not be improved by reducing the background level. Also for short half-life products, the flux must be high enough to keep irradiation times samll, or flux/time details must be known.

We conclude that TL resulting from phosphor activation is a feasible means for detecting fast neutrons. For $CaSO_4$, application would be facilitated by fabrication of single-piece dosimeters since handling radioative powders is at best inconvenient.

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RESUMO

A técnica de ativação pode ser usada para detetar qualquer radiação que é capaz de ativar um dosímetro termoluminescente. Neste trabalho, $CaSO_4$:Dy (Harshaw) foi irradiado no reator do Instituto de Energia Atômica. O dosímetro é envolto numa folha de Cd quando o efeito de nêutrons térmicos deve ser eliminado afim de detetar nêutrons rápidos. A reação $^{32}S(n,p)^{32}P$ com uma secção de choque efetiva de 265 mb na região de 2,7 MeV produz ^{32}P radioativo, que induz no decorrer da desintegração (com meia vida de 343 horas), TL no $CaSO_4$:Dy. O efeito da irradiação inicial, principalmente de raios gama é eliminado por recozimento em temperatura suficientemente alta (600°C por 30 min) logo após a irradiação. A TL auto induzida é uma medida da fluência de nêutrons rápidos. Na região de energia de reação (E $\gtrsim$ 2,7 MeV), a estimativa dá para a fluência mais baixa detetável, um valor correspondente a um rad de Kerma no tecido vivo.

RÉSUMÉ

La technique d'activation peut être utilisée pour la detection de toutes les radiations capables d'activer un dosimètre thermoluminescent. Dans ce travail, $CaSO_4$:Dy (Harshaw) fut irradié dans le reacteur de l'Institut d'Energie Atomique. Le dosimètre est enveloppé dans une feuille de Cd pour éliminer l'effet des neutrons thermiques pour la dosimetrie des neutrons rapides. La reaction $^{32}S(n,p)^{32}P$ avec une section de choc de 265 mb autour de 2,7 MeV, produit ^{32}P radioactif qu'induit, pendant sa desintegration (avec une periode de desintegration de 343 heures), TL dans $CaSO_4$:Dy. L'effet de l'irradiation initiale, principalement de rayonnement gamma, est eliminé par un recuit à une temperature suffisamment haute (600°C pendant 30 minutes) imédiatement aprés l'irradiation. La TL auto-induite est une mésure de la fluence des neutrons rapides. Dans la région d'énergie de la réaction (E $\gtrsim$ 2,7 MeV), l'évaluation de la plus petite fluence detectable donne une valeur correspondante a un rad de Kerma dans le tissu.

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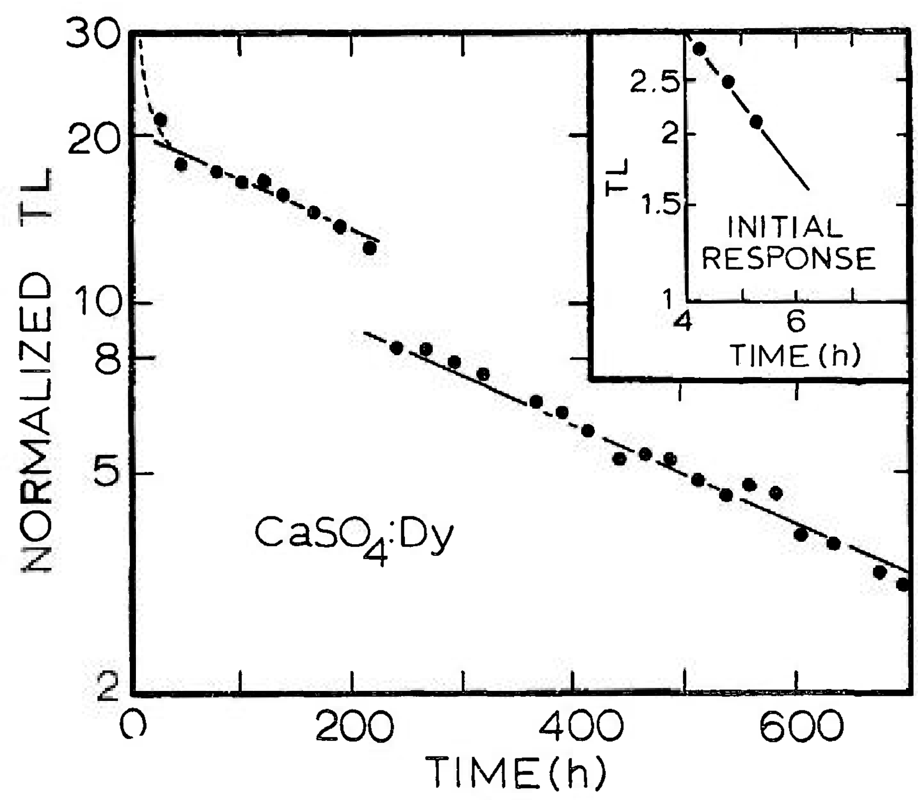

FIGURE CAPTION

Fig. 1. - The daily TL from self-irradiation as a function of time after irradiation in the reactor. The solid lines represent decay with the 343h half-life of ^{32}P. The discontinuity results from annealing the sample 30 min at 600°C (see text). Since the self-irradiation times are not always identical, each TL reading is divided by $\exp(\Delta t/495)-1$. Insert: The initial TL due to 30 min self-irradiations. The solid line represents decay with the 2.32 h half-life of ^{165}Dy.