THERMAL AND EPITHERMAL NEUTRON DOSIMETRY BY INDUCED ACTIVATION IN COLD-PRESSED PELLETS OF TL PHOSPHOR-MIXTURE*

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ABSTRACT Measurement of thermal and epithermal neutron exposures have been attempted using CaF_2 and $CaSO_4$ TL phosphors cold-pressed into pellets after mixing with carefully chosen fluxing agents which have appreciable cross sections in this neutron energy range. Epithermal and thermal exposures have been arbitrarily distinguished by the difference obtained with and without cadmium filters covering the pellets; the TL measured in each case was due to the "self-irradiation" by the induced activity after the "prompt dose" has been removed immediately after theirradiation by a suitable thermal annealing procedure. Some of the mixtures investigated are : (CaF_2 + KBr) and ($CaSO_4$ + KBr) for thermal neutron dosimetry by the induced bromine activity and (CaF_2 + Dy_2O_3 + KCl) and ($CaSO_4$ + Dy_2O_3 + KCl) for thermal and epithermal neutron dosimetry by the induced dysprosium activity. The pellets made of the above mixtures have been found to be convenient for repeated use in radiation dosimetry and their neutron sensitivities have been evaluated for exposures to 400 μgm ^{252}Cf source and for irradiation in the core as well as the beam hole of IEAR-1, a swimming pool reactor. The minimum detectable fluences are estimated to be of the order of 10^8 n.cm^{-2} for thermal neutrons and 10^{10} n.cm^{-2} for epithermal neutrons. The paper describes the optimum mixture compositions and the perfomance characteristics on the pellet dosimeters.

Introduction

The present work was undertaken as an extension of two earlier studies viz. a proposal to use powder mixtures of TL phosphor and KBr and TL phosphor and

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Dy_2O_3 for thermal and epithermal neutrons, respectively (1) and another, to use cold pressed pellets of TL phosphors mixed with KCl in routine dosimetry (2). It was decided to study the performance of TL phosphor-KBr powder mixture in thermal neutron detection after cold pressing them into pellets while in the case of the TL phosphor-Dy_2O_3 powder mixture it was decided to add KCl and cold press them into pellets for use in epithermal neutron detection.

The general principles underlying the detection of thermal and fast n-activation induced TL have been described earlier /3,4). Considerations regarding the half-lives of products involved and the choice of optimum time lag between erasure of prompt dose-induced TL signal and the reading of activation induced TL, in the case of presently taken up phosphors mixtures have been described elsewhere (1,5).

Experimental

The TL phosphors used in the present study were $CaSO_4$(Dy) and natural CaF_2. While the former was prepared in the laboratory by the usual co-precipitation and evaporation technique, the latter was a fluorite mineral of Brazilian origin which was cleaned and crushed before use. Spec-pure Dy_2O_3 powder was used as the neutron activator while laboratory reagent grade KCl was used as the flux to make cold-pressed pellets (2). In another set of experiments the TL phosphors were individually mixed with laboratory reagent grade KBr which acted both as the neutron activator and the flux to make the pellet dosimeters.

The weight of either kind of pellets was 180 mg with the discs having approximately 1 cm diameter and 1 mm thickness. The optimum compositions of the TL phosphor + KBr mixture and TL phosphor + Dy_2O_3 mixtures (with KCl weight fixed at 90 mg) were experimentally found out both for the prompt gamma dose TL response and the n-activation induced TL response. While the former exhibited a monotonously decreasing response with decreasing weight of the TL phosphor, the latter gave a peak response for an optimum composition of the TL phosphor - n activator mixture (fig.1 & 2). All other experiments were conduced for this optimum phosphor concentration in the pellets.

Unacceptable inhomogenity occurred in the distribution of the n-activator agent in the pellets pf $CaSO_4$:Dy-KBr and CaF_2-Dy_2O_3-KCl which resulted in a wide scatter of the data.These are hence not included in the present paper. Incidentally, the TL transmission characteristics have been found to be the best for the combinations of CaF_2+KBr and $CaSO_4$+Dy_2O_3+KCl, from photoluminescence studies (6).

Irradiations were carried out near the core of IEAR-I swimming pool reactor using a pneumatic drive for activation periods of the order of seconds; at one

of the beam holes of IEAR-I, for activation periods of the order of minutes and inside water at 2.5 cm from a 400 μg ^{252}Cf source for activation periods of the order of hours. The thermal fluxes at these locations were of the order of 10^{11}, 10^{7} and 10^{6} with Cd ratios of about 17, 13 and 6 respectively as determined by fold foil activation experiments.

Significant half-lives of the beta-active activation products of KBr are 4.5 hours and 3.6 hours and that in the case of Dy_2O_3 is 2.4 hours. For the sake of convenience and standardisation purposes, both the type of dosimeters after each activation exposure, were anneled immediately (maximum delay was about 15 mins and could be easily aceounted for) at 600°C for 20 min , theself irradiation from the decaying activity was allowed for a 24 hours period and then activity induced TL was read. While for Dy_2O_3, this 24 hour self-irradiation period resulted in the maximum obtainable sensitivity, for KBr this gave only slightly less than the maximum obtainable sensitivity (24 hours correspond to more than 7 half-lives of the decaying Dy activity; nearly 6 half-lives of one of the decays of Br which was produced with 6 = 13.5 barns and less than one half-life for the other decay of Br whose σ= 3.3 barns).

Results and Discussions

It is interesting to note the difference in the shapes of the two curves presented in figs. 1 & 2 for the optimum mixture compositions of CaF_2+KBr and $CaSO_4$:Dy+Dy_2O_3(+KCl). The TL emission spectra are different as also the optical absorption spectra of the fluxing agents of KBr and KCl; besides in the latter case, the presence of Dy_2O_3 as a third component in the pellet makes the average distance between the beta emitter and the TL phosphor grain greater than in the case of KBr pellets for the activation induced TL responses. The difference in the shapes of the response curves could very well be due to the combined effects of these factors.

Fig. 3 presents the results obtained for the n-irradiations done on Caf_2 + KBr pellet dosimeters. These dosimeters gave less than 10% responses after Cd filtration and these have been subtracted in the plots. It is worth mentioning here that a line of linearity has been obtained for an entire fluence range of 10^{8} - 10^{14} n cm $^{-2}$ from various sources on a log-log scale but not with a slope of unity. Fig.4 presents the results obtained for the n-irradiations done on cadmium covered KCl pellet dosimeters; here an unit slope has been obtained for short time irradiation in the reactor and a sublinear plot has been obtained for the longer irradiations done with ^{252}Cf source. It is known that in activation experiments one can assume fairly

linear activation for exposure periods less than 1/3 or 1/4 times the half-life of the activation product. This has been experimentally verified to be true for the case of activation induced TL (4). Thus even in fig. 4 (b) an unit slope line can be assumed upto 40 min of irradiation and then a gradual fall towards satuation. The case of CaF_2 + Kbr is complicated because of simultaneous activities with different half-lives being present and scatter in the results are more for longer irradiation periods as seen in fig. 3 . Besides, KBr is fairly TL sensitive and has a peak at larger doses more or less coinciding with the CaF_2 dosimetry peak. This may be the reason for the slight supralinearity obtained. Nevertheless the log-log linear plot can still be useful in dosimetry.

Conclusions

It is obvious that the $CaSO_4$+Dy_2O_3+KCl pellet dosimeters can be used in pairs - with and without Cd filters to monitor efficiently both the thermal and epithermal neutrons. (Dy thermal activation cross section may be hundred times more than that of Br). The advantage with KBr is only that in this case one does not need a separate binding agent to make the pellet dosimeters. Extrapolation from the present results indicate that thermal neutron fluences of the order of 10^{8}n cm^{-2} and epithermal fluences (of a light water moderated fission spectrum) of the order of 10^{10}n cm^{-2} could be easily detected by these pellet dosimeters. While the chief advantage of this activation induced TL dosimetry lies in its freedom from the omnipresent gamma background in the neutron field, the main disadvantage still remains that this can be used only for very short exposures and requires an immediate erasive thermal treatment and hence may be useful only in area monitoring.

Although these pellet dosimeters are almost 1000 times less sensitive than the powder mixtures in activation dosimetry (1,5), an unique possibility could be envisaged with these pellet dosimeters: the beta activity induced by the neutron exposure can be absolutely measured from a calibration for the K-40 beta induced self irradiation of the dosimeter. Such a measurement can lead to the estimations of the neutron flux accurately just as in any foil irradiation and counting measurements, provided some kind of equivalence could be established between the K-40 beta activity and the Br/Dy beta activity as the case may be. Thus these pellet dosimeters can serve as even secondary standards in neutron flux measurements, the technique being lot easier than the absolute foil counting technique. This is being investigated now.

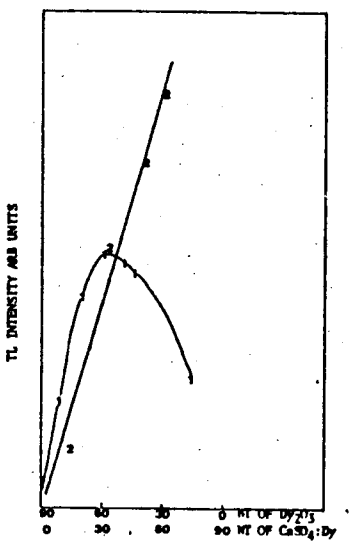

Fig. 1

Optimum mixture composition of $CaSO_4$ (Dy) and Dy_2O_3 for a fixed weight of the KCl flux in the pellet dosimeter.

1: prompt gamma response

2: n-activation induced response

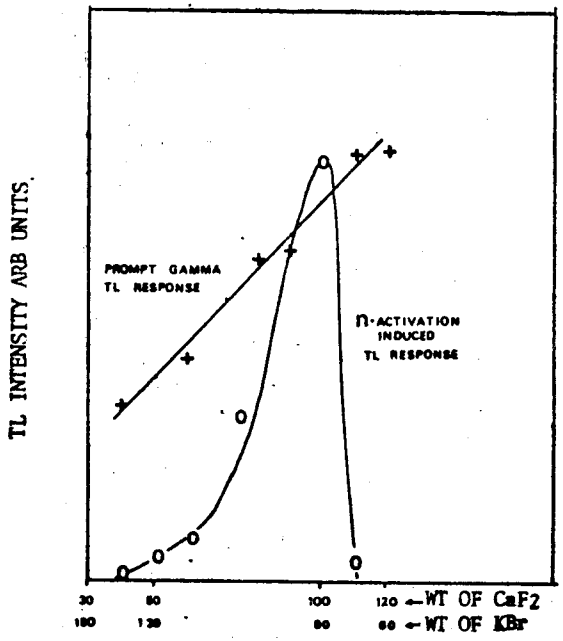

Fig. 2

Optimum mixture composition of natural CaF and KBr in the pellet dosimeter

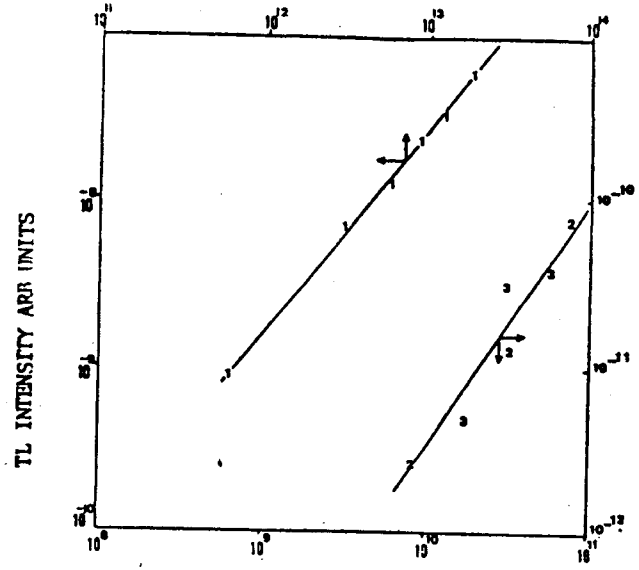

Fig. 3

Thermal-neutron-activation induced TL outputs of CaF_2 +KBr mixtures for various neutron irradiations

1: Reactor core irradiations

2: Beam Hole irradiations

3: Cf source exposure inside water

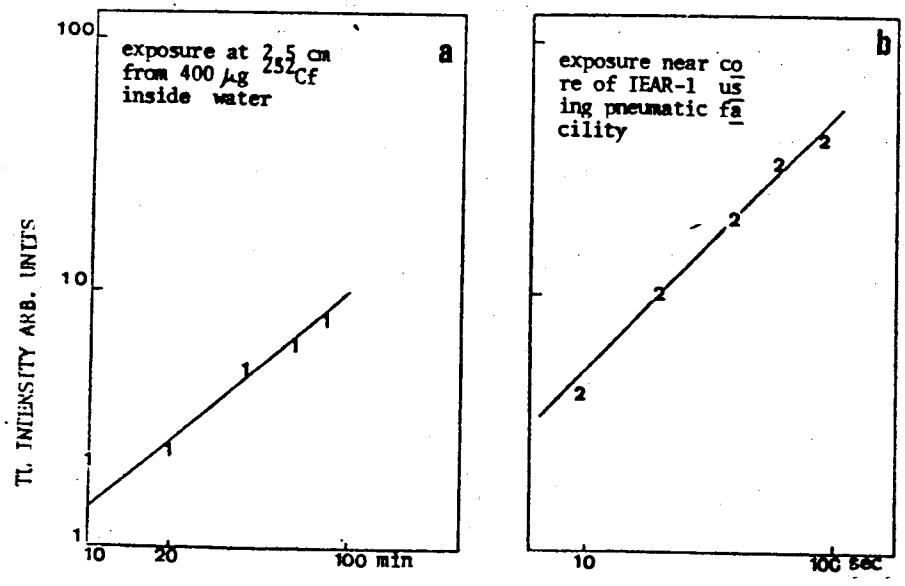

Fig. 4 Nepith activation induced TL outputs of Cd covered $CaSO_4$ + Dy_2O_3 + KCl pellets for various neutron irradiations

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