radiation effects, 1970, Vol 5, pp. 99–102

OBSERVATION OF THERMOLUMINESCENCE INDUCED BY FISSION FRAGMENTS

K. V. ETTINGER, S. A. DURRANI AND C. CHRISTODOULIDES
Department of Physics, University of Birmingham, Birmingham 15, England

Thermoluminescence induced in CaF_2 powder by fission fragments emanating from a uranium foil bombarded by fast neutrons has been measured as a function of neutron fluence. A linear relationship between the glow produced and the fast neutron fluence between 5×10^{10} and 6.5×10^{11} n/cm^2 has been obtained, thus establishing the feasibility of the use of this method for fast-neutron dosimetry. A limitation of the method is that, if the fissile foil is not separated from the phosphor after irradiation, the TL produced by the α-disintegration of ^{238}U may eventually mask the fission-induced TL.

1. INTRODUCTION

Thermoluminescence (TL) has been successfully employed in the past in the dosimetry of γ-rays and charged particles, and, more recently, of neutrons. As far as the detection of fast neutrons by means of the TL induced by them is concerned, the proton recoil technique has been employed[1,2] as an intermediary, but with moderate success only. A search of the literature has failed to bring to light instances of TL induced by fission being used for the detection of fast neutrons. An attempt has therefore been made to study the feasibility of such an approach.

2. EXPERIMENTAL PROCEDURE

Calcium fluoride was considered to be a good choice as a phosphor for fast neutron measurement *via* their fission yield, since it has been reported[3] that CaF_2 exhibits no detectable direct sensitivity to fast neutrons. Moreover, CaF_2 has one of the highest light yields known[4], so that a relatively simple TL reader can be used for the measurements.

The fissile material employed was in the form of a foil, which was brought into contact with TL powder only during the time of irradiation by neutrons. Figure 1 shows the irradiation container, being a light-tight holder inside which the TL phosphor (CaF_2) and the fissile material (^{238}U) are pressed together. The ^{238}U is in the form of a thick (0.4 mm) foil, while the phosphor is deposited as a layer of free powder glued either to cellulose acetate or to an aluminium backing by means of 'Zapon' lacquer. After irradiation, the phosphor is removed from the backing by means of a solvent. The best solvent was found to be hexane; but acetone, when not exposed to light, also gave good results. Some solvents, e.g. trichloroethylene, exhibit considerable quenching of the thermoluminescence—possibly because of the impurities present in them. It was further found that the handling of the irradiated phosphor in the form of a suspension reduces the effects of spurious (i.e. non-radiogenic) TL. In order to obtain good reproducibility of the quantity of the phosphor used, as well as to ensure the uniformity of spreading of the sample on the heating strip, it was found best to dispense, by means of a micrometer syringe, an accurate amount of the phosphor which had been made into a slurry or a suspension.

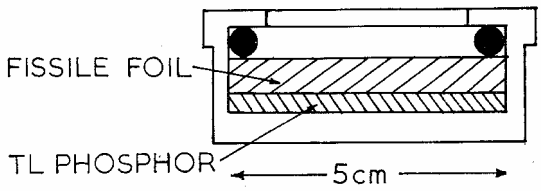

FIG. 1. Schematic diagram of the TL-fission detector (not drawn to scale). A ^{238}U foil (0.4 mm thick) is held in contact with the CaF_2 phosphor powder (glued as a free layer to an acetate or aluminium backing) during the neutron irradiation. The composite detector is housed in a light-tight container. After the irradiation, the two halves are separated and an accurate amount of the phosphor is used for TL readout.

The lower limit of the neutron flux that can be detected by this method is set, in theory, by the rate of α-decay of the fissile material. The ranges of α-particles from the decay of ^{238}U, and those of main fission products, are in fact very similar, being typically ~ 13 mg/cm^2 for fission products and ~ 16 mg/cm^2 for α-particles, in uranium metal (or ~ 5 mg/cm^2 in CaF_2 powder). The corresponding

linear ranges are $\sim 8\,\mu$m in uranium and $\sim 15\,\mu$m in CaF_2 for both α's and fission products. It is thus clear that no useful enhancement of the fission/alpha ratio is possible on the basis of difference in ranges. Simple calculation shows that the flux of fast neutrons at which the rate of fission equals that of alpha disintegration of ^{238}U is about 5×10^6 n/cm^2 sec. There remains, however, a great excess of energy deposited by fission fragments in comparison to that deposited by α-particles (assuming comparable TL efficiencies). In actual fact, the limit of detectable fluxes is set by the sensitivity of the phosphor and the quality of TL reader. We found experimentally that fluxes of the order of 10^7 n/cm^2 sec are easily detectable, without the need for cooling the photomultiplier used in the TL reader.

The highest fluence that can be recorded was not determined because of practical limitations. Predicting from the known characteristics of CaF_2 when exposed to γ-rays, the limit of linearity of CaF_2 response is probably at about 10^4 rads,[5] which corresponds to a fluence of $\sim 10^{13}$ n/cm^2.

As a neutron source we use the $^7Li(p, n)^7Be$ reaction, employing 10 MeV protons from our 60-inch cyclotron. The total yield of our target was determined to be $(1.8 \pm 0.2) \times 10^{10}$ n/μA (M. C. Scott, private communication, 1969). At the position where irradiations were carried out, the fast neutron flux was measured to be 8.9×10^7 n/cm^2 sec. The number of fission events affecting the TL powder is calculated to be about 6×10^2 fissions/cm^2 sec.

The irradiated samples are protected from the slow neutron flux, and also to some extent from the γ-rays from the target, by using suitable shielding materials. The neutron spectrum emerging from the target in the forward direction is shown in Figure 2. The figure also shows the ^{238}U fission cross section as a function of the neutron energy. The fast neutron flux is monitored by a thorium-lined fission chamber. Because of uncertainties in the determination of the fast neutron spectrum, it was not possible to calculate the precise dose received by the powder. It is also uncertain whether the secondary-electron equilibrium resulting from the fission-fragment ionization had been attained. Following the standard method[6] of calculating the dose imparted by the elastic scattering of fast neutrons, and taking into account the energy deposited by fission fragments in the CaF_2, the total dose rate in CaF_2 powder is calculated to be approximately 270 mrad/sec at 6 μA of proton

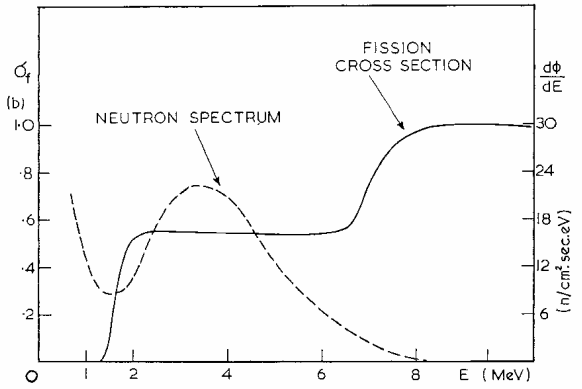

FIG. 2. The neutron energy spectrum (flux per eV) incident at the detector, resulting from the $^7Li(p, n)^7Be$ reaction in a thick target exposed to a 6 μA beam of 10 MeV protons from the Nuffield Cyclotron; and the ^{238}U fission cross section $\sigma_f(E)$, in barns, over the energy-range of interest.

current from the cyclotron. Of this, the fission-fragment dose constitutes over 90 per cent ($\sim$255 mrad/sec).

The results of measurements of the TL produced in the phosphor, using the ^{238}U foil, are shown in Figure 3, up to a fluence of 6.5×10^{11} neutrons/cm^2.

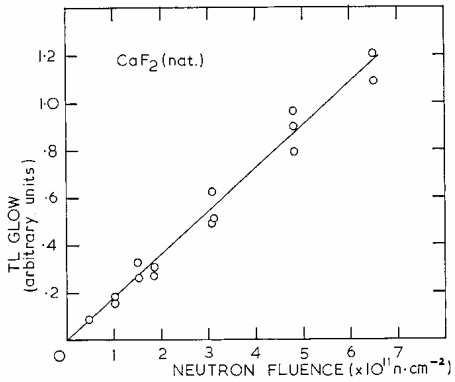

FIG. 3. Total TL glow (in arbitrary units) produced in the CaF_2 phosphor held in contact with the ^{238}U fissile foil, shown as a function of the neutron fluence (i.e. time-integrated flux) incident upon the detector. A linear relationship is observed between 5×10^{11} and 6.5×10^{10} n/cm^2.

A control experiment in which the CaF_2 powder was exposed to the same neutron beam, but with the fission foil separated from the TL phosphor, gave a TL glow ~ 6 per cent of that produced when the fission foil was in contact. This background response includes the direct interaction of fast neutrons, as well as the effects of the γ-rays present.

Some of the slowed-down neutrons might also contribute to this background. The other source of background TL, viz. that produced by the α-particles resulting from ^{238}U disintegration, has already been discussed above. It sets the final limit to the detectable fluxes of neutrons, but provided that the uranium foil is separated from the powder after bombardment with neutrons, the effect is negligible for fluxes of the order of 10^7 n/cm^2 sec or above.

3. SOURCES OF INTERFERENCE

Fission cross sections are relatively low for fast neutrons within the energy range of interest (see Figure 2). Fortunately, however, not many competing nuclear reactions can take place in CaF_2 with fast neutrons; and those that do, have low cross sections as well. We have calculated the approximate values of reaction rates for these competing processes in addition to fission itself. These are summarized in Table I. Values for (n, γ) reactions with slow neutrons (which have comparatively low fluxes) are assumed to be negligible. It will be seen that reactions $^{40}Ca(n, p)^{40}K$ and $^{40}Ca(n, \alpha)^{37}A$ have rates comparable with fission; but fortunately the energy deposited in the phosphor from these reactions is many times smaller than that deposited by fission fragments, because of the much greater energy carried by the latter.

These competing reactions, nevertheless, do increase the total TL response of the powder somewhat, and must, therefore, be taken into account for accurate dosimetric work.

TABLE I

Reaction rates $\eta \cdot \int_{E_{min}}^{E_{max}} \sigma_\gamma(E)\, \phi(E)\, dE$ calculated for the total experimental fast-neutron flux 8.9×10^8 n/cm^2 sec; η being the natural abundance.

Reaction	Weighted reaction rate (events per nucleus per sec)†	Product half-life
$^{238}U(n, f)$	4.1×10^{-17}	—
$^{40}Ca(n, p)^{40}K$	2.1×10^{-17}	1.3×10^9y
$^{40}Ca(n, \alpha)^{37}A$	1.6×10^{-17}	35d
$^{42}Ca(n, p)^{42}K$	4.2×19^{-21}	12.5h
$^{42}Ca(n, \alpha)^{39}A$	4.1×10^{-21}	265y

† 1. The reaction rates have been calculated by using the neutron flux per unit energy, $\phi(E)$, resulting from the cyclotron, as shown in Figure 2, and the corresponding microscopic cross section, $\sigma_\gamma(E)$, at energy E. The rates have been weighted according to the natural abundance, η, of the isotope concerned.

2. Reaction rates in F are completely negligible. So are those in ^{44}Ca.

The response to γ-rays from the target and from the room background, and response to slow neutrons present, were not separately measured. This response is estimated to be about 5 per cent of that due to fission in view of results of Fremlin (private communication, 1969), who estimates, under more adverse conditions, the contribution of γ-rays to be about 10 per cent of the total response to fast neutrons, using the same neutron target. It cannot, however, be ruled out that in certain circumstances (e.g. in a reactor environment) the γ-ray contribution to the total TL yield may become unacceptable.

4. CONCLUSION

It is possible to measure fast neutron fluxes larger than 5×10^6 n/cm^2 sec by using a TL phosphor in contact with a layer of fissile material. The disadvantage of the method is the alpha emission from the fissile material which might mask the TL produced by fission if the powder remained in contact with the fissile material for a prolonged period of time. It seems, therefore, that this method would be less suitable for routine neutron flux measurements if the composite detector described above had to be kept as a permanent monitor (i.e. if the separations of the fissile foil from the phosphor were not possible). Nevertheless the technique is of potential use in the calibration of neutron fluxes. Moreover, if the ratio of the TL efficiencies of α-particles and of fission fragments can be determined accurately (e.g. by varying the time of irradiation while keeping the fluence constant), allowance can be made for the glow caused by the α-decay of the fission source, so that prolonged use of such a monitor would be permissible. Alternatively, a fissile foil highly enriched in ^{235}U and exposed to thermal neutrons should prove very much more favourable in its fission-induced TL in comparison with the α-induced background and other competing (n, γ) reactions. We plan to perform such an experiment.

ACKNOWLEDGEMENTS

The authors wish to thank Professor J. H. Fremlin for his support of this work and for helpful discussion, and Dr. M. C. Scott for supplying the neutron spectrum data which enabled us to calculate most of the reaction-rate values quoted in the paper.

REFERENCES

1. R. A. Facey, *Trans. Am. Nucl. Soc.*, **11** (1), 415–16 (1968).
2. V. V. Kuz'min, Ch. B. Lushchik, F. A. Savikhin, A. D. Sokolov and I. V. Yaek, *At. Energ.* (*USSR*), **22,** 482–88 (June 1967). (In Russian.)
3. J. Spaepen, *Phys. Rev. Letters*, **1,** 281 (1958).
4. Z. D. Spurný, *Phys. Med. Biol.*, **13,** 465–66 (1968).
5. J. R. Cameron, N. Suntharalingam, and G. N. Kenney, *Thermoluminescence Dosimetry* (University of Wisconsin Press, Madison, Wis., 1968), p. 47.
6. *N.B.S. Handbook 85, Physical Aspects of Irradiation* (U.S. Government Printing Office, Washington, D.C., 1964).

Received 26 March 1970