THE MEASUREMENT OF GAMMA-RAY ENERGY DEPOSITION IN THE ZERO POWER FAST REACTOR ZEBRA WITH Li\textsuperscript{7}F THERMOLUMINESCENT DOSIMETERS

by

Alan D Knipe

Introduction

1 Gamma-rays are a significant heat source in a fast nuclear reactor since their absorption contributes approximately ten per cent to the total reactor energy. The optimisation of cooling and shielding requirements demands reliable data and methods of calculation in order to predict accurately energy deposition and penetration. A stringent test of the adequacy of data and methods is provided by comparison with experimental results from zero power fast reactor assemblies. Such a comparison was carried out in the ZEBRA NZB and NZC assemblies during the KOZANT program (1). A complete dose-mapping of NZB, which was a large plutonium core without singularities, was made with a zirconium-walled ionisation chamber and a zirconium-walled solid-state cavity dosimeter (SSCD) incorporating Li\textsuperscript{7}F thermoluminescent dosimeters (TLD). The NZC assembly contained tantalum and boron carbide mock-up control rods. TLD samples were used to measure gamma-ray energy deposition directly within the absorber pins.

2 The TLD "cavity", introduced to probe the charged particle spectrum generated by photon interactions in the "wall" medium, constitutes a discontinuity which affects the equilibrium spectrum. The relationship between the energy deposition in the cavity and the wall is a function of photon energy, cavity size and composition of the media. The form of this relationship is a major problem when measurements are made within different materials in a reactor. A study of this problem and details of the KOZANT gamma-ray energy deposition measurements are given in Reference (2); this paper summarises the interpretation of thermoluminescent output of Li\textsuperscript{7}F TLD following irradiation of a fast reactor.

The TLD Technique

3 High sensitivity Li\textsuperscript{7}F (0.007% Li\textsuperscript{6} and 99.993% Li\textsuperscript{7}), manufactured by the Harshaw Chemical Company under the name TLD-700 in the form of a solid rod 1 mm x 1 mm x 6 mm, was chosen for the ZEBRA work. Thermoluminescent lithium fluoride has been investigated by many workers (3-6). The Li\textsuperscript{7}F crystals are found to operate over a wide dose-range; exhibit high photon sensitivity; relatively low neutron sensitivity and the reproducibility by manufacturers is established. Provided care is exercised when handling the crystals, accurate, reproducible results can be achieved, and the dosimeters can be utilised for several experiments provided they are suitably annealed. The post-irradiation annealing process used during this work was to heat them for one hour at 400°C, followed by sixteen hours at 80°C. The crystals were "read out" using an Isotopes/Con-Rad Model 7100 TLD Reader. In this particular equipment the crystals are placed on a nichrome planchet which, when loaded into the reader, becomes part of the electrical heating circuit. The light output was measured with a photomultiplier tube operating in the current mode. The constant rate of heating was determined by the current through the planchet. This was chosen such that when the glow curve output had returned to zero (or at worst one-third up the main peak) the heater current would switch off. A stable light source was substituted for the planchet at frequent intervals to check drift in the photomultiplier gain and all read-out counts were normalised to the standard light source reading. Prior to read-out, the irradiated crystals were cleaned in chloroform and alcohol.

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The TLD were calibrated in a known Co60 field, the length of irradiation giving the desired dose as LiF TLD are dose-rate independent. The crystals were surrounded by a lithium fluoride wall of equilibrium thickness during exposure. The response per rad for TLD-Co60 can be considered constant (7,8) in the fast reactor photon energy range (100 keV to 8 MeV). The dose response curve (ie dose vs integrated light output) is linear up to approximately 200 rad(LiF). The response in the supralinear region at doses above this is a function of gamma-ray energy and therefore reactor power and irradiation times were chosen to give doses in the linear region of the dose response curve. Annealing alters the dose response of the TLD and therefore batch calibrations were essential after each experiment. As the ZEBRA core temperature was below 50°C there was no change of response due to irradiation at an elevated temperature. Following irradiation, there is a slow rate of loss of stored energy ("fading" of the TL signal). During the MOZART project the problem of uncertainties in the rate of fading was overcome by carrying out the read-out process at a fixed time (seven days) after irradiation.

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In TLD-700 the neutron reactions of importance are the Li$^6$ (n,$\alpha$)t reaction and elastic scattering of the Li and F nuclei. Based on a response of 0.15 for the alpha-particles and tritons relative to Co60 gamma-rays, the energy deposition from the Li$^6$ (n,$\alpha$)t reaction was calculated to be less than 0.1% in the HZB core and blanket regions, increasing to about a per cent in the outer reflector regions. The major contribution to the TL signal was caused by the Li$^7$ and F recoils. The sensitivity to the recoil nuclei in TLD as a function of energy was obtained from the work of Tanzka and Furuta; the most recent data are given in Reference (9). The correction for the neutron response in the HZB core was typically 15% and the associated uncertainty was estimated to be 1% (1 sd).

The Cavity Correction

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Standard calculational methods for predicting gamma-ray energy deposition in reactors do not account for the migration of secondary charged particles - the energy transferred from a photon to an electron or positron being assumed to be deposited at the site of interaction. If the dimensions of a region of interest are comparable with the range of the charged particles (as in the case of the TLD), this assumption is not valid. Photon-electron transport codes exist (eg 10) but they are difficult to adapt for the efficient solving of the cavity problem. The approach adopted for the ZEBRA work was to write a Monte Carlo electron tracking program, separate from the gamma-ray codes, that would operate from calculated photon spectra. Computed spectra consist only of source, Compton and two-quantum annihilation photons. The binding energy loss in the photoelectric effect is ignored and bremsstrahlung losses are not included. It is also assumed that two-quanta annihilation of the pair-production positron only takes place when the particle has come to rest, and annihilation with the emission of a single photon is ignored. With the exception of bremsstrahlung losses at high energies these assumptions have a negligible effect in most heating and shielding calculations.

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The basic concept of the electron tracking program (PROCIDE) was the calculation of the energy deposited in the cavity whilst containing the wall medium ($E_w$) and the energy deposited in the cavity whilst containing the detecting medium ($E_d$). The resulting ratio ($E_w/E_d$) gave the required cavity correction. Thus, for the ZEBRA work, the dose-rate in a material of interest was calculated using standard methods and PROCIDE related this value to the experimental result.

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The energy deposited in a cavity arises from charged particles generated within the cavity itself and from the "wall contribution volume"; the boundaries of which are defined by the maximum range of the most energetic particles generated by photon interactions. From the input photon spectrum PROCIDE generates
charged particle source spectra for the wall medium and detecting medium (eg LiF). These spectra are a function of the photon spectrum and the relevant macroscopic photon interaction cross-sections. For a photoelectric interaction it is assumed that all the photon energy is transferred to the electron in order to be consistent with standard gamma-ray programs. For the Compton effect the recoil electron energy is taken from the distribution given by Evans (12). The Bethe-Heitler pair-production distribution is used to obtain the electron and positron kinetic energies (13). Particles are generated from the appropriate secondary source spectrum with a uniform spatial distribution and an isotropic angular distribution.

Because of the immense number of collisions a charged particle undergoes in slowing down, it is not possible to represent every individual collision as in a gamma-ray Monte Carlo program. For example, a 0.5 MeV electron in gold requires approximately \(1.7 \times 10^5\) collisions to lose half its energy. It is therefore necessary to group many steps of the real physical history into a single step to give a "condensed" history. The average energy deposition for each step in PROCEED is calculated from the continuous-slowing-down model. The main inelastic collision loss is given by the Bethe equation using the formulation of Michael (14), no allowance being made for the radiative loss. In PROCEED the positrons created during pair-production are treated as electrons. Elastic collision with a nucleus predominates in the history of a charge particle and at the end of each step a polar direction (\(\theta\)) is chosen from a multi-angle scattering distribution. The exact determination of the distribution function has been carried out in the small-angle approximation by Snyder and Scott, and Molière. The data and method used in PROCEED were obtained from a review article on small-angle scattering by Scott (15) and a paper by Bethe (16) on Molière's theory. The later paper discusses the relationship with an exact theory by Goudsmit and Saunderson which is valid for any angle by means of an expansion in Legendre polynomials. PROCEED uses the Molière theory to compute the total scattering angle distribution and extends the theory to include large angles by multiplying the distribution by a factor \((\theta/\sin\theta)^2\) as suggested by Bethe. Inelastic collisions were accounted for by extra terms in the Molière expressions. The angular distributions of multiply scattered electrons calculated with the Molière theory showed good agreement with the general Goudsmit-Saunderson theory. The azimuthal angle for the Monte Carlo is sampled from an isotropic distribution. A logarithmic step size is chosen in order that the average angular multiple scattering deflection per step remains approximately constant. Step sizes are calculated by PROCEED to fulfil the Molière theory conditions. The relative number of photon interactions in the cavity and wall contribution is obtained by maintaining equilibrium between the number of particles entering and leaving the cavity when it contains the wall medium. The number of interactions in the cavity when it contains the detecting medium may then be calculated from the relevant reaction rates.

A benchmark experiment was designed to investigate the cavity correction in the absence of complications due to the reactor environment. A 22 Ci Co60 source was housed in the centre of an iron cube of side 600 mm. A 10 mm diameter access hole ran 100 mm below the source plane into which various test materials (LiF, C, Fe, BaO2, Ta) were loaded; some of these wall materials were actual control rods and structural test pins used for energy deposition measurements within the ZEBRA core. The LiF crystals were located at the centre of the wall materials. An irradiation time of 30 minutes was chosen to restrict the TL light output to the linear region of the dose response curve. Each experiment was repeated several times, giving a standard error of better than 1\% in most cases. The crystals were readout seven days after irradiation using the procedures previously described.

The photon spectra and dose-rates at the centres of the various test materials were calculated with the Monte Carlo code HORSE (17). A 27-group P5 multigroup
cross-section set was generated from the UK Nuclear Data Library, which is based on the data of Hubbell (18). Difficulties in these calculations due to the use of the Legendre expansions for the angle of scatter and the point-flux estimator are discussed in Reference (2). The calculated spectra were used as input to PROCEED. Additional data required were the cavity dimensions, material descriptions, atomic weights and the average excitation-ionisation potentials. Photon interaction cross-sections were again obtained from the Hubbell data.

The results from the Iron Block Benchmark are summarised in Table 1 - statistics for the NORSE calculations were + 3% and the PROCEED calculations + 2%

<table>
<thead>
<tr>
<th>Wall</th>
<th>Experimental Result (rad hr⁻¹)</th>
<th>NORSE Corrected Ext Result (rad hr⁻¹)</th>
<th>NORSE Calculation (rad hr⁻¹)</th>
<th>C/E</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiF</td>
<td>187.7</td>
<td>190.0</td>
<td>190.0</td>
<td>1.01</td>
</tr>
<tr>
<td>Fe</td>
<td>179.0</td>
<td>243.0</td>
<td>243.0</td>
<td>1.02</td>
</tr>
<tr>
<td>Ta</td>
<td>159.7</td>
<td>239.1</td>
<td>239.1</td>
<td>0.99</td>
</tr>
<tr>
<td>Eu₂O₃</td>
<td>203.0</td>
<td>349.0</td>
<td>349.0</td>
<td>1.01</td>
</tr>
</tbody>
</table>

The C/E values in Table 1 show excellent agreement between the PROCEED corrected measurements and the NORSE calculations, and it was concluded that the benchmark validated the TLD technique and the cavity Monte Carlo code in the Co60 spectrum energy range.

An experimental study of the cavity correction with monoenergetic sources in the energy range 0.145 MeV to 8 MeV has been carried out by Van Prooyen (19). The quantity \( \varepsilon_c \) was measured for LiF surrounded by materials of varying atomic number and \( \varepsilon_w \) calculated assuming that the absorbed dose was only due to primary photon interactions. Burlin (20) has derived an expression to calculate the cavity correction for the situation where cavity dimensions are comparable with the electron range. This was attained by combining the dose due to electrons produced within the cavity, and externally produced electrons, by means of a weighting factor which was a function of the maximum electron energy. In general, Van Prooyen's experiments are in agreement with predictions from the Burlin theory and PROCEED. A significant discrepancy, however, exists between experiment (and the Burlin theory) and PROCEED results in the high energy region for low-Z cavities surrounded by high-Z walls. Results from the Monte Carlo calculation in this energy region in tantalum are at least 20% greater than the values obtained from the Burlin theory. If multiple scattering is neglected and particles are assumed not to deviate over their entire path length the PROCEED values agree with those from Burlin's theory. The Monte Carlo method should be the more rigorous approach to the cavity problem and further work is essential to establish the cavity correction in the high energy region where PROCEED does not corroborate the Burlin theory.

Measurements within ZEBRA

A Solid State Cavity Dosimeter (SSCD) was devised capable of making an axial or radial scan in ZEBRA while at power during a single irradiation. The SSD...
consisted essentially of a zirconium rod, 9 mm diameter and 1.2 m in length with twenty 1 mm x 1 mm x 6 mm cavities at 50 mm intervals down the longitudinal axis of the rod. A further twelve cavities, identical to the above, were equally spaced between the first and third cavities in order to study fine structure which results from the plate composition of the ZEBRA elements. A typical irradiation time at full power was 15 minutes. Axial scans in the NZB assembly were made in normal ZEBRA elements containing a 24 mm diameter hole which extended to a depth just below the core centre plane. The radial scans were made in an 11 mm diameter stainless steel tube located horizontally along the major axis of the core centre plane. TLD were also incorporated in stainless steel ZEBRA plates (6 crystals per plate), to allow accurate mapping of the rapid dose change in the breeder region. For the measurements within the Ta and B4C absorber rods, demountable pins with axial locating holes were used, the length of the pellet determining the axial position of the TLD. The control rod measurements required that the crystals were loaded before reactor start-up. The irradiations were therefore carried out at a lower power (usually 10% FP) in order to give a long irradiation time compared with the build-up time to power and post-shutdown irradiation. Details of the MCZART calculations and the various corrections necessary (eg non-saturation of fission product activity) are given in Reference (2).

Figure 1 compares a typical SSCD scan with Monte Carlo calculations. The standard deviation on the measurements is 10%; the calibration and reproducibility accounting for 5% and the various corrections for the remainder. The cavity correction was approximately 10% in the core region - this decreased slightly in the blanket and then increased rapidly in the reflector regions. In the tantalum control rod experiments at the NZC core centre the cavity correction was approximately 20%, and agreement with calculation for both Ta and B4C was better than 7%. Recent measurements have been made in the benchmark test materials in a sub-assembly at the ZEBRA core centre; reproducibility to within 1% has been achieved between TLD measurements made on different days. With more accurate calibrations and an improvement in the uncertainties of the corrections it should be possible to attain a standard deviation of 5% on TLD measurements.

Conclusions

The MCZART gamma-ray energy deposition experiments have demonstrated the practicability of Li7F TLD measurements in a zero power fast reactor environment. Accurate, reproducible results can be achieved provided the crystals are handled with care and sufficient attention to detail is taken during the read-out stage. The TLD, when incorporated in the SSCD, gave a useful method of obtaining dose-rate scans while at power, and the crystals proved suitable for measuring the energy deposition directly within materials. Discrepancies between MCZART and the monoenergetic source measurements in the MeV region must be studied further due to the central role of the cavity correction in interpreting TLD-type measurements. Methods of obtaining a more accurate evaluation of the fast neutron correction also require investigation.

Reactor Physics Division
UKAEA
AEE Winfrith
Dorchester
Dorset
United Kingdom

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COMPARISON OF MONTE CARLO CALCULATION OF
ABSORBED DOSE-RATE AND TLD MEASUREMENTS
ALONG MAJOR AXIS OF ASSEMBLY MZB (2)

FIGURE 1
REFERENCES


14 Charged-Particle Interactions
   Chapter 4 of Reference 11
   H Bichsel

15 The Theory of Small-Angle Multiple Scattering of Fast Charged Particles
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   W T Scott

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   H A Bethe

17 The STOP Code - A Multigroup Neutron and Gamma-Ray Monte Carlo Transport
   Code
   ORNL-4585 (1970)

18 Photon Cross-Sections, Attenuation Coefficients and Energy Absorption
   Coefficients from 10 keV to 100 GeV
   NSRDS-NBS 29 (1969)
   J H Hubbell

19 High Energy Gamma-Ray Energy Deposition Within Shielding Materials
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20 A General Theory of Cavity Ionization
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- 8 -
ATTENTION OF MR DAVID FISCHER, IAEA

RE COORDINATION MTG 14-15 NOV. IN RESPONSE TO YOUR TLEX OF 29 OCT TO WILLIAMS AND FURTHER TO MY LETTER OF 26 OCT, YOUR SUGGESTED AGENDA ITEMS ARE AGREED. WE PROPOSE SEVERAL ADDITIONAL ITEMS, YIELING A CONSOLIDATED LIST AS FOLLOWS:

1. GENERAL COORDINATION INCLUDING PROCEDURES ON SYMPOSIA AND CONFERENCES, PRINCIPAL STAFF, PRESENT AND PROJECTED, GEN STRATEGY FOR COMPLEMENTARITY IN AREAS SUCH AS SAFETY, RAD PROTECTION

2. SAFETY, INCL STOCKHOLM CONFERENCE, TMI FOLLOWUP PROGRAMS, NUSS, TRANSPORTATION SAFETY (INCL YOUR BSB), COMPILATION ON NUCL ACCIDENTS (YOUR 3), BEFAST (YOUR 8)

3. RAD PROT (INCL CCC) MILL TAILINGS (INCL YOUR SYMPOSIUM AND YOUR DDD), EMERGENCY PLANNING (INCL SCHEDULE FOR IAEA REPORT), GASEOUS EFFLUENTS (INCL SYMPOSIUM), ICRP-26 IMPLEMENTATION, OCCUPATIONAL EXPOSURE (AAA)

4. WASTE MGT, SEA DUMPING, MONITORING, PACKAGING (HHH), SYMPOSIUM ON IMPACT OF RAD NUCL ON MARINE ENV, HIGH LEVEL WASTE RAD PROT PHILOSOPHY, LEGAL STUDY, NUCLIDE MIGRATION DATA BANK, NE; GEO DISPOSAL PROJECTS, TRANSFORMATION (EEE), D AND D (FFF), SEA BED (GGG)

5. FUEL CYCLE, JOINT GROUPS ON SUPPLY (YOUR 4), DEMAND (6), UREPI PROS AND NET, EXPLORATION R AND D (5), EXTRACTION, FUTURE INVOLVEMENT IN INSTITUTIONAL MATTERS

6. NUCLEAR SCIENCES, CODE AVAILABILITY, DISTRIBUTION STATISTICS (YOUR 9), DATA AND CODE COMPATIBILITY (INCL E.G. URANIUM DEMAND), NEACRP OBSERVERS (YOUR7), 1981 SHORN MTG

I PROPOSE TO CONTACT YOU EARLY IN THE WEEK TO DISCUSS SPECIFIC DETAILS OF ARRANGEMENTS. MANY THANKS FOR YOUR EFFORTS

WILLIAM H HANNUM
DEPUTY DIRECTOR GENERAL
NEA

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AEN NEA PARIS