AAEC/TM347



# AUSTRALIAN ATOMIC ENERGY COMMISSION

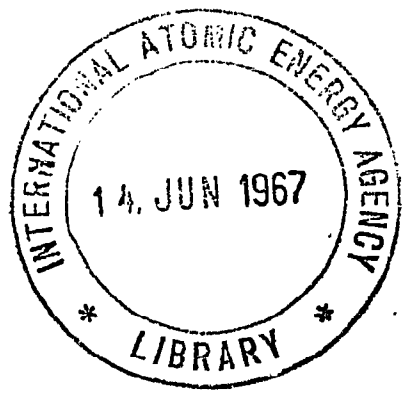
## RESEARCH ESTABLISHMENT

## LUCAS HEIGHTS

# RADIATION FIELD CHARACTERISTICS AND IRRADIATION TECHNIQUES FOR GAMMA IRRADIATION FACILITIES USING SPENT FUEL ELEMENTS FROM THE REACTOR HIFAR

by

B.M. O'LEARY




October 1966

AUSTRALIAN ATOMIC ENERGY COMMISSION
RESEARCH ESTABLISHMENT
LUCAS HEIGHTS

# RADIATION FIELD CHARACTERISTICS AND IRRADIATION TECHNIQUES FOR GAMMA IRRADIATION FACILITIES USING SPENT FUEL ELEMENTS FROM THE REACTOR HIFAR

by

B.M. O'LEARY

## ABSTRACT

Experimental and service gamma irradiations of various small volume or bulk materials can be carried out at Lucas Heights by using an array of spent fuel elements from the reactor HIFAR in an irradiation pond. Details of the gamma radiation fields available are given and the irradiation techniques are described. These include experimental irradiations at low, ambient, and above-ambient temperatures. Geometrical factors which affect the dose rate are taken into account to ensure that the materials receive the specified dose.

CONTENTS

## 1. INTRODUCTION

Spent fuel elements from the A.A.E.C.'s reactor HIFAR are transported to an irradiation pond and used as underwater sources for experimental and service gamma irradiations. This "Spent Fuel Element Gamma Irradiation Facility" has been described elsewhere (O'Leary 1966). The elements are located in an array of fixed vertical tubes at the bottom of the pond with the centres spaced 12 inches apart. Most irradiations are carried out by lowering a 10 inch diameter irradiation container into the array so that the material packed inside the container receives radiation from the adjacent elements.

Fuel elements may be of either box or hollow cylindrical type and, before irradiation, contained either 115 or 150 g of highly enriched uranium. The activity of a spent fuel element varies according to its type, its reactor history, and its cooling time. Dose rates of up to 2 megarads per hour have been obtained inside a 10 inch diameter irradiation container but up to 5 megarads/hour is possible by using a special 1.5 inch diameter irradiation container fitted inside a hollow fuel element.

This report gives details of the radiation fields obtained in the facility and sets out the methods used to irradiate material to specified doses.

## 2. RADIATION FIELD CHARACTERISTICS

### 2.1 Methods of Measuring Radiation Fields

The most important quantity to be measured is the maximum dose rate in a standard air-filled irradiation container in the various positions in the spent fuel element array. This is most simply measured by an ion chamber placed inside a special water-tight irradiation container which has a vented lid and flexible polyethylene tubes running from the vents to a point above the water surface. The electrical leads from the ion chamber pass through the plastic tubes to a voltage supply and microammeter on a movable bridge over the pond. A calibration graph (based on chemical dosimetry) is used to convert ion chamber current to the maximum air dose rate in a standard irradiation container and from this, effective dose rates for the irradiation of various samples can be calculated as described in Section 3.

Chemical dosemeters are also used for routine measurements of dose rates but they are most useful for monitoring doses absorbed by irradiated material and for plotting radiation field distributions. The Fricke ferrous sulphate dosemeter (Draganic et al. 1961) is used for doses in the range 3,000 to 45,000 rads. The solution is usually contained in polyethylene capsules 6 cm long and 1.7 cm in diameter. The increase in optical density of the solution at a wavelength of

3040 Å due to conversion of ferrous ion to ferric ion is used as a measure of the absorbed dose. For measurement of doses in the range 1 to 20 megarads the ceric sulphate dosemeter is used. The solution is contained in Pyrex glass bottles 3 cm high and 1.8 cm in diameter. The reduction in optical density measured at 3200 Å due to conversion of ceric ion to cerous ion under irradiation is proportional to the absorbed dose.

To plot radiation fields, dosemeters are simply located at points of interest in the irradiation container and exposed for a suitable time. Examples are given in the next section.

2.2 Radiation Field Distribution in Various Media

It is essential to know the field distribution inside an irradiation container when it is empty and also when it is filled with materials of various densities, in order to calculate exposure times for the irradiation of materials. The rig used to measure dose rate variations consists essentially of two vertical plastic tubes which run the length of the irradiation container, one at the centre and one at the wall. Chemical dosemeters fit snugly inside the tubes in two columns. Figure 1 shows typical results for an empty irradiation container and one filled with materials of various densities. For simplicity,only the fields along the central axis are shown.

Field variations in a plane across an irradiation container through the point of maximum dose rate are of particular interest. Chemical dosemeters were used to determine this for a single element (Figure 2a). Figure 2b is drawn from Figure 2a and shows the dose rate variations resulting from the superimposed fields of four equal fuel elements in the four adjacent positions. Even in this ideal situation the air dose rate at points on the circumference varies from about 14 per cent. less than the maximum dose rate on the centre axis to about 10 per cent. more, so a container must be rotated during an irradiation. For an air-filled irradiation container the effect of rotation is to make the mean dose rate at the side about 2 per cent. less than at the centre. For most purposes it is sufficient to turn the container through $\frac{3}{8}$ of a revolution by hand at half the exposure time. For more precise experimental irradiations a motor placed inside the container rotates the material continuously.

2.3 Decay Rate

The dose rate decay has been determined for particular single elements (Figures 3 and 4) by measuring the maximum dose rate in the 10 inch diameter air-filled irradiation container due to the adjacent element. Measurements were made



at various cooling times. The dose rate varied from about 400,000 rads/hour after cooling for 1 week to about 6,000 rads/hour after cooling for a year. Typical decay rates were 6-7 per cent. per day after cooling a week, 2 per cent. per day after 50 days, and less than 1 per cent. per day after 1 year.

2.4 Energy Consideration

The dominant contributors to the gamma dose rate over the period of use of the spent fuel element sources are the isotopes Zr-95 and its daughter Nb-95 (half-lives 65 days and 35 days respectively). These emit gamma rays with energies around 0.75 MeV. Calculations from graphs given by Moteff (1954) show that for fission products formed by 1000 hours of reactor operation, about 50 per cent. of the gamma energy emitted after 10 days' shutdown is due to Zr-95 and Nb-95. This percentage increases to about 85 per cent. after 100 days' cooling time. The weighted mean gamma energy decreases from about 0.86 MeV at 10 days to about 0.74 MeV at 100 days, and remains at about this level for at least a further 6 months, which covers the period of use of the elements as radiation sources. The decrease over the first two months is mainly due to the more rapid decay of fission products with energies grouped around 1.65 MeV.

Estimates of the mean gamma energy of radiation which penetrates to the inside of an irradiation container are complicated by the geometry of the facilities. In general, radiation from the important Zr-95/Nb-95 isotopes will be degraded somewhat by Compton scattering, mainly in the water surrounding the elements. On the other hand water-shielding will greatly reduce the contributions from low energy isotopes and make high energy emitters (for example Ce-144/Pr-144) proportionately more important.

Changes in mean gamma energy over the first few months' cooling time could lead to significant variations in the gamma attenuation in irradiated material when the same material is irradiated at different cooling times. This in turn could lead to errors in the estimation of the absorbed dose at the centre of the material. To test whether a significant change in attenuation rate with cooling times does in fact occur, comparisons were made of the dose rate at the centre of an empty container with the dose rate at the centre of a water-filled container. No really significant difference in the ratio of these dose rates was observed over the period of use of the fuel elements. Attenuation rates in other low atomic number materials were also measured at various cooling times and found to be approximately constant for a particular material.

2.5 Neutron Flux

Some neutrons are produced by ($\gamma$, n) reactions between photons with energy greater than 2.2 MeV and deuterium in the water. La-140 with a 2.5 MeV emission is the main source of these reactions. La-140 (half-life 40.2 h) is a daughter of Ba-140 which has a half-life of 12.8 days so that the highest neutron flux will result from spent fuel elements within the first few weeks after removal from HIFAR. Measurements of neutron flux by activation of gold foil have given values of less than $10^3$ n $cm^{-2}$ $sec^{-1}$.

## 3. IRRADIATION OF MATERIAL

Exposure times for the irradiation of material are based on ferrous sulphate dosimetry. For gamma radiation from spent fuel elements energy deposition will predominantly be due to Compton scattering, particularly as the materials commonly irradiated have low atomic numbers. Thus the energy absorbed by a particular material will depend on its electron density. Most substances irradiated have an electron density fairly close to that of the Fricke dosemeter ($3.334 \times 10^{23}$ electrons/g) but in some cases it may be necessary to apply corrections according to the electron density ratio of the material and the Fricke dosemeter.

3.1 Small Volume Experimental Irradiations

Small volume experimental irradiations usually include single small samples or a number of test tubes of chemicals.

Single samples are located centrally on a stand with their volume equally spaced above and below the point of maximum dose rate. The smaller the sample the lower is the dose variation over its volume. A minimum dose, rather than a mean dose, is usually specified. Table 1 indicates the minimum dose rates for small samples of various sizes and densities expressed as a percentage of the prevailing maximum dose rate in the air-filled irradiation container. The approximate maximum overdose is also given. These results were obtained by trial irradiations of samples with small chemical dosemeters inserted. For best results, and wherever practicable, trial irradiations are carried out with dosemeters inserted in a duplicate of the actual sample.

Test tube samples are usually spaced apart in holes in plastic foam blocks, thus reducing self shielding by the tubes and fixing the geometry for repeated irradiations. Again trial irradiations with ferrous sulphate dosemeters are carried out beforehand. An average dose rate is worked out and expressed as a percentage of the maximum air dose rate so that for similar experiments at later dates the average dose rate and the required exposure time can be found by taking



the same percentage of the prevailing maximum dose rate in the air-filled irradiated container. When the actual irradiation is carried out, one or more ferrous sulphate or ceric sulphate dosemeters are added to monitor the dose. For long irradiation exposures a correction is applied for the decay during the exposure time.

The stand used for supporting the sample usually consists of a plastic foam cake on a light aluminium stand. This minimises sample shielding from below. Backscatter from such a stand is small (about 2 per cent.).

Rotation of the samples is usually required to counter radiation field asymmetry.

Experimental irradiation at low temperatures may be carried out by immersing the samples in a liquid nitrogen or dry ice bath in a vacuum flask in the irradiation container*. In these cases the irradiation container is fitted with a vented lid with a spiralling polyethylene tube attached. The gas escapes up the tube to the atmosphere, thus avoiding pressure build-up. When samples are being irradiated in liquid nitrogen the air is first purged from the irradiation container by nitrogen gas so that ozone does not condense in the flask during the irradiation. (B.P. of ozone = -112°C, M.P. = -192°C). Ozone formation from oxygen impurity in the liquid nitrogen is more difficult to combat. Flasks are of stainless steel and are kept well topped up to avoid concentration of ozone.

Irradiations at above-ambient temperatures may be carried out by means of a thermostatically controlled, heated oil bath in a vacuum flask in the irradiation container. The bath is stirred by a stream of air which enters (and empties) through the vented lid connections. Electrical leads also enter through the polyethylene tube connected to the lid. A hot water jacket connected by tubes with a thermostatically controlled water bath at floor level has also been used successfully to provide temperatures up to 75°C. This system avoids electrical leads and the risk of insulation break-down under irradiation.

The vented irradiation container may also be used to circulate air or nitrogen past a sample under irradiation.

---

*For experiments requiring more flexible control of low temperature a Co-60 refrigerated facility is available.

3.2 Large Volume Irradiations using Inversion of Material

(a) Material packed into two standard size packages: Most bulk material for irradiation is packed into packages 9.5 inches in diameter and 24 inches high. Two such packages are loaded in an irradiation container, one below the point of maximum dose rate and the other above it. After half the total exposure time their positions are reversed.

Generally speaking for bulk irradiations of this kind a minimum dose is specified. Table 2 shows the minimum effective dose rate (that is the dose rate averaged over the total exposure time for the part of the package which receives the lowest dose) for materials of various densities. This is given as a percentage of the maximum dose rate along the centre axis of the air-filled irradiation container. Knowing the maximum air dose rate and the weight of the material in the standard packages, the minimum effective dose rate can be obtained from the Table and the total exposure time calculated. For high doses a correction should be applied for decay of dose rate during the exposure time. Table 2 also gives values for the approximate overdose to be expected. Values for the minimum effective dose rate and the overdose, in Table 2, were obtained by irradiating standard packages containing material of various densities and with chemical dosemeters inserted. Additional values were obtained by interpolation.

(b) Material packed into one standard size package: For a single canister of material the same procedure is followed except that a stand 24 inches high is used to support the package during one half of the exposure time. Experiments have shown that the minimum effective dose rate is the same as it would be for two packages irradiated together, but the overdose is increased. For the most common case of a single package with a bulk relative density of 0.5 the maximum overdose is increased by about 5 per cent.

(c) Material packed into one or two non-standard size packages: It is occasionally necessary to irradiate material supplied in packages of various lengths and diameters. According to the accuracy required, minimum effective dose rates can be estimated from Figure 1 or a trial run with a similar package containing dosemeters.

3.3 Irradiations in Hollow Fuel Elements

In special cases where a very high dose or dose rate is required for small samples, use is made of the intense field in the core of a spent hollow fuel element. A 1.5 inch diameter stainless steel irradiation container is located in the element. A typical field along the axis of the container as measured by ferrous sulphate dosemeters is shown in Figure 5. Maximum dose rates of up to



$5 \times 10^6$ rads/hour are obtained. The maximum dose rate is about ten times the maximum dose rate in the 10 inch diameter air-filled irradiation container due to this one element.

## 4. SOME GEOMETRICAL FACTORS AFFECTING THE DOSE RATE IN A FACILITY

### 4.1 Orientation of Box-Type Spent Fuel Elements with Respect to a Facility

Box-type fuel elements are approximately square in section with the parallel fuel plates slightly convex towards one face. Measurements of the dose rate at the centre of a 10 inch irradiation container due to a single box type element are approximately 5 per cent. higher if the convex face is directed towards the container instead of the concave face or the other two sides.

### 4.2 Changing Position of Fuel Elements Nearby

The main contribution to the dose rate in an irradiation container comes from the elements in the array which are immediately adjacent to the container. There are usually four of these. Other elements contribute to a lesser extent. For a fresh highly active fuel element located in the nearest of the neighbouring (non-adjacent) tubes, the contribution to the maximum dose rate at the centre of an air-filled irradiation container is about 40,000 rads/hour. In an extreme case where four old fuel elements are adjacent to an irradiation container, the effect of putting a fresh spent fuel element in a neighbouring tube could be to double the original dose rate. Care is taken to move elements only when they are not affecting any irradiation in progress.

### 4.3 Movement of Irradiation Containers in Neighbouring Facilities

If an irradiation container is in position in the central facility of an array of spent fuel elements, the placing of irradiation containers in adjacent facilities is likely to reduce the shielding between nearby elements and the first irradiation container, leading to an increase in dose rate. The effect is most pronounced when the nearby containers contain mainly air space and when very active elements surround the central four elements. If one air-filled container is interposed between the central facility and two very active fuel elements, the increase in the maximum air dose rate in the central container can be up to 40,000 rads/hour. For an array of spent fuel elements of approximately equal activities, an air-filled irradiation container loaded into a neighbouring facility is likely to increase the maximum dose rate in an adjacent air-filled container by about 3 to 4 per cent. Movement of irradiation containers in and out of facilities is therefore controlled so that significant errors in irradiation dose exposure are not made.

## 5. CONCLUDING REMARKS

The spent fuel element gamma irradiation facilities are now in routine operation. Radiation fields and handling techniques have been established so that most irradiations can be undertaken on a routine basis. However some research irradiations still need special treatment, as for example when dosimetry problems are involved or the temperature of samples must be controlled during irradiation.

The fuel element irradiation facilities have been mainly used for experimental work, nearly three-quarters of the irradiations carried out in them being for research purposes, either for the A.A.E.C. or for external research organisations. As experimental facilities they provide relatively uniform radiation fields and a wide range of useful dose rates. A disadvantage is the rapid decay rate of spent fuel elements which means that considerable effort is devoted to routine dose rate measurements.

## 6. REFERENCES

Draganic, I.G., Holm, M.W., and Maul, F.E. (1961). - Laboratory manual for some high level chemical dosimeters. Riso Report No.22, Danish Atomic Energy Commission.

Moteff, J. (1954). - Miscellaneous data for shielding calculations. APEX - 176.

O'Leary, B.M. (1966). - A gamma irradiation facility using spent fuel elements. Atomic Energy in Australia, 9 (2) : 7 - 11.

TABLE 1

DOSE RATES FOR GAMMA IRRADIATION OF SMALL SAMPLES

A minimum dose is usually specified. The minimum effective dose rate for samples of various sizes and densities is expressed as a percentage of the maximum dose rate along the central axis of an air-filled irradiation container.

| Relative Density | Height (in.) | Diam. (in.) | Minimum Dose Rate as % of Max. Air Dose Rate | Approx. Percent. Over-dose |
|---|---|---|---|---|
| 0.2 | 6 | 2 | 94 | 5 |
| | | 4 | 93 | 6 |
| | | 6 | 92 | 7 |
| | | 8 | 91 | 8 |
| | | 10 | 90 | 8 |
| | 12 | 2 | 87 | 14 |
| | | 4 | 86 | 13 |
| | | 6 | 86 | 12 |
| | | 8 | 85 | 11 |
| | | 10 | 85 | 11 |
| 0.5 | 6 | 2 | 93 | 6 |
| | | 4 | 90 | 8 |
| | | 6 | 87 | 10 |
| | | 8 | 84 | 12 |
| | | 10 | 81 | 14 |
| | 12 | 2 | 84 | 16 |
| | | 4 | 81 | 18 |
| | | 6 | 77 | 19 |
| | | 8 | 74 | 21 |
| | | 10 | 71 | 23 |
| 0.75 | 6 | 2 | 91 | 6 |
| | | 4 | 88 | 8 |
| | | 6 | 84 | 9 |
| | | 8 | 80 | 11 |
| | | 10 | 76 | 13 |
| 0.75 | 12 | 2 | 83 | 17 |
| | | 4 | 78 | 19 |
| | | 6 | 74 | 22 |
| | | 8 | 69 | 24 |
| | | 10 | 65 | 28 |
| 1.00 | 6 | 2 | 89 | 9 |
| | | 4 | 82 | 13 |
| | | 6 | 76 | 18 |
| | | 8 | 69 | 24 |
| | | 10 | 62 | 31 |
| | 12 | 2 | 81 | 18 |
| | | 4 | 75 | 22 |
| | | 6 | 69 | 27 |
| | | 8 | 63 | 32 |
| | | 10 | 57 | 35 |
| 1.50 | 6 | 2 | 88 | 10 |
| | | 4 | 80 | 16 |
| | | 6 | 72 | 23 |
| | | 8 | 65 | 32 |
| | | 10 | 57 | 43 |
| | 12 | 2 | 81 | 17 |
| | | 4 | 74 | 20 |
| | | 6 | 68 | 23 |
| | | 8 | 61 | 27 |
| | | 10 | 55 | 32 |

TABLE 2

DOSE RATES FOR GAMMA IRRADIATION OF MATERIAL IN TWO STANDARD PACKAGES

Two standard size (height 24 inch, diameter 9.5 inch), equal-weight packages are irradiated at once, their positions being reversed at half the exposure time. The minimum effective dose rate for packages of various weights is expressed as a percentage of the maximum dose rate in the empty irradiation container.

| Weight of Each Canister (lb) | Bulk Relative Density | Minimum Effective Dose Rate as per cent. of Max. Air Dose Rate | Approximate Maximum Overdose (per cent.) |
|---|---|---|---|
| 10 | 0.16 | 49 | < 10 |
| 15 | 0.24 | 48 | < 10 |
| 20 | 0.32 | 46 | < 10 |
| 25 | 0.40 | 45 | < 10 |
| 30 | 0.48 | 43 | < 10 |
| 35 | 0.56 | 42 | < 10 |
| 40 | 0.64 | 40 | < 10 |
| 48 | 0.72 | 38 | 16 |
| 56 | 0.80 | 37 | 16 |
| 62.5 | 1.00 | 33 | 25 |

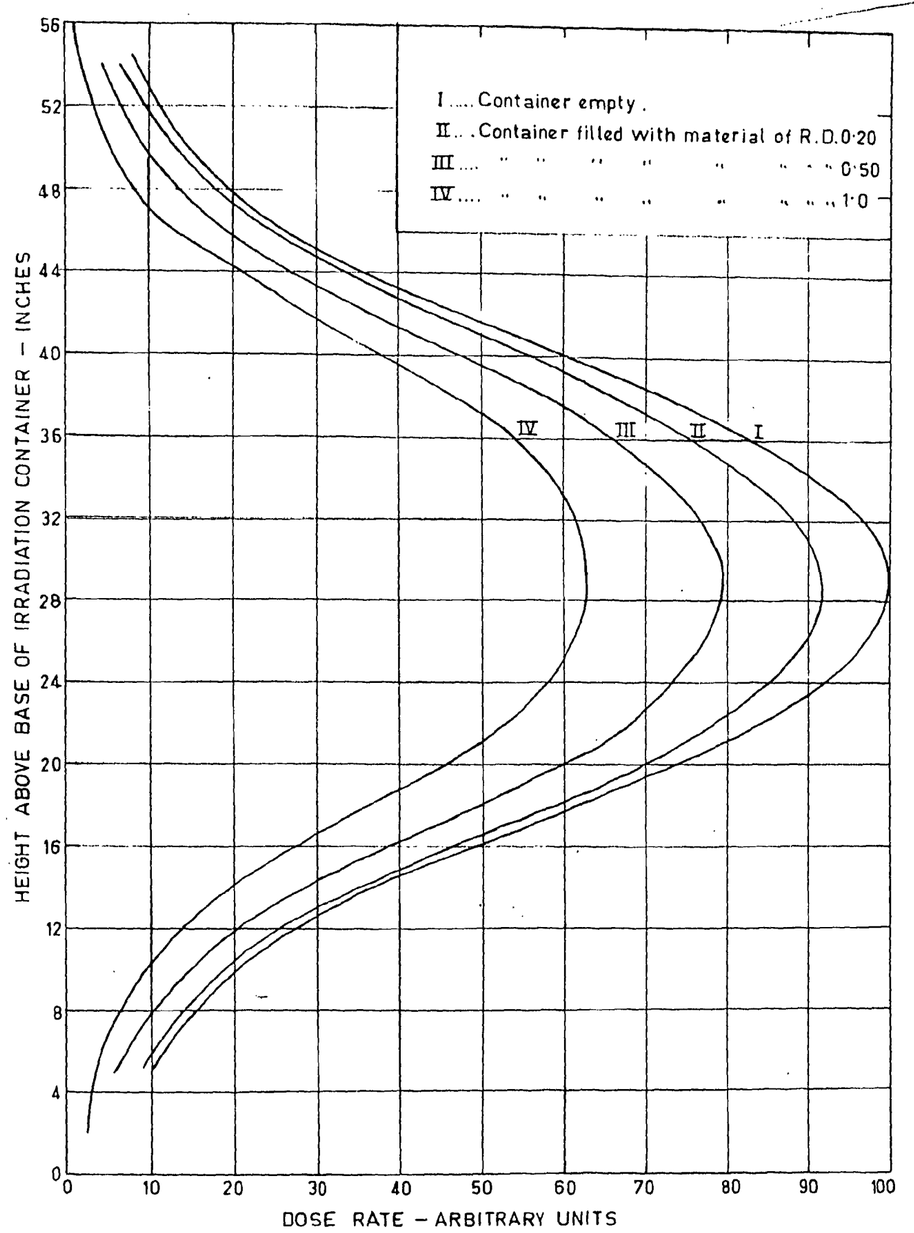
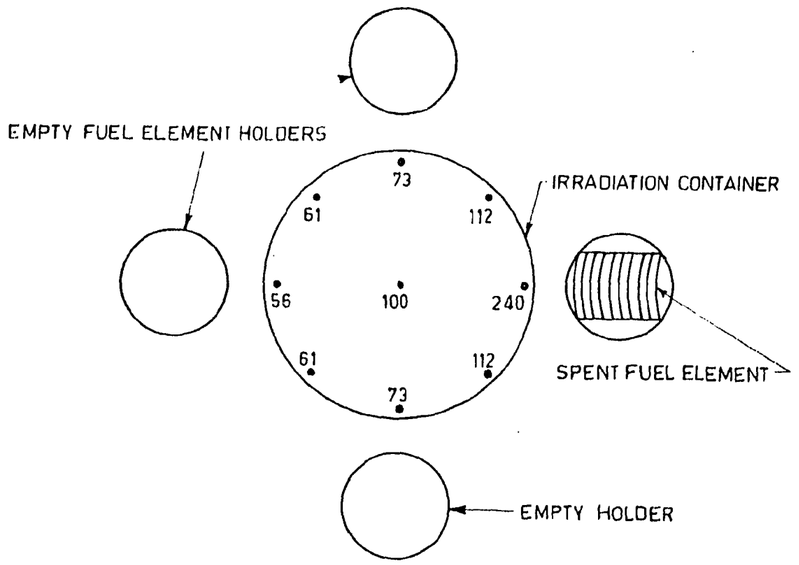
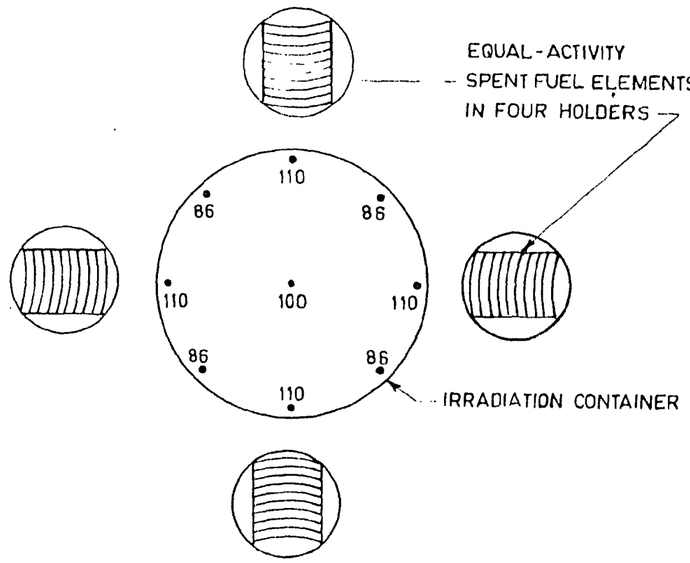



**FIGURE 1. RADIATION FIELD DISTRIBUTION ALONG THE AXIS OF AN IRRADIATION CONTAINER FILLED WITH MATERIAL OF VARIOUS BULK RELATIVE DENSITIES**

P1001




(a) ONE ADJACENT SPENT FUEL ELEMENT




(b) FOUR ADJACENT EQUAL-ACTIVITY SPENT FUEL ELEMENTS

**FIGURE 2. TYPICAL DOSE-RATE DISTRIBUTION IN A HORIZONTAL PLANE THROUGH THE POINT OF MAXIMUM DOSE-RATE IN AN AIR-FILLED IRRADIATION CONTAINER**

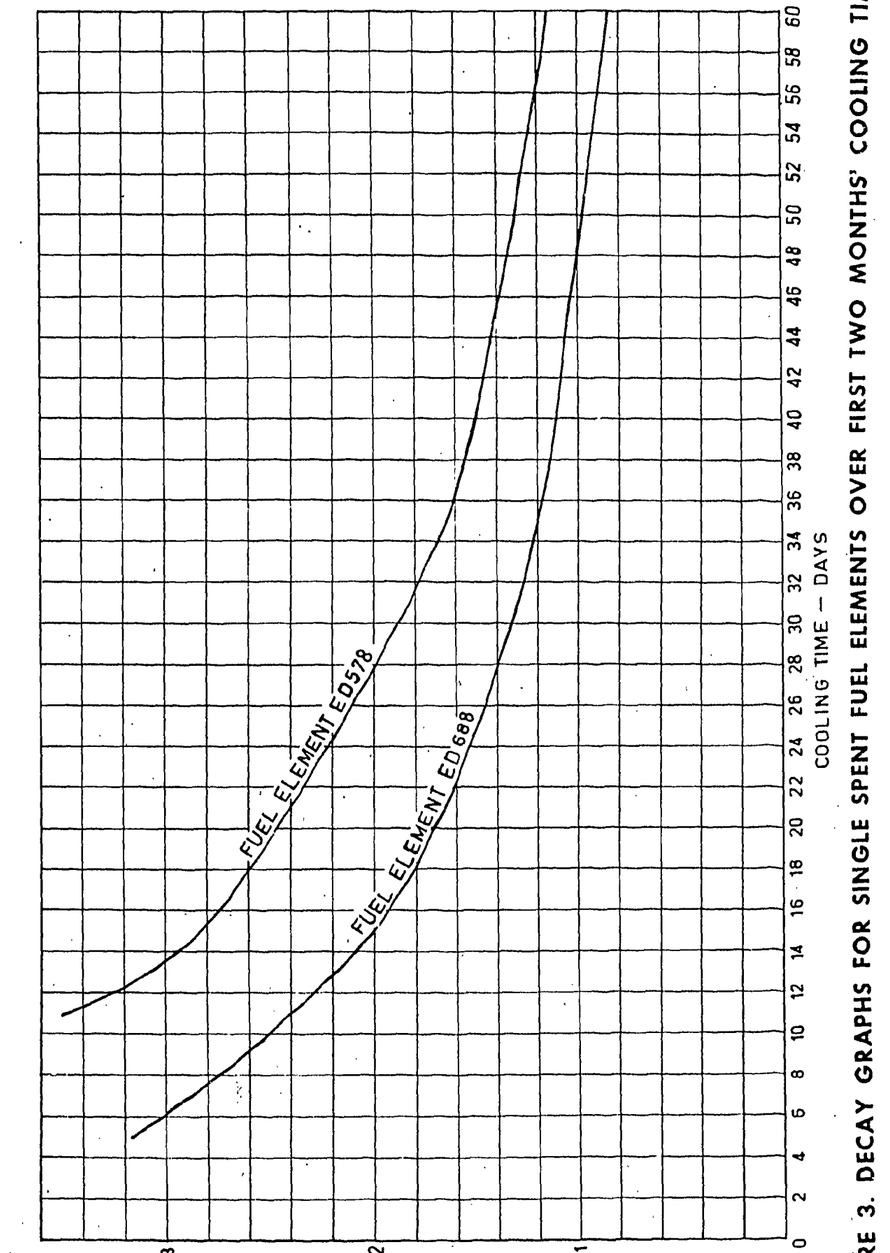



**FIGURE 3. DECAY GRAPHS FOR SINGLE SPENT FUEL ELEMENTS OVER FIRST TWO MONTHS' COOLING TIME**

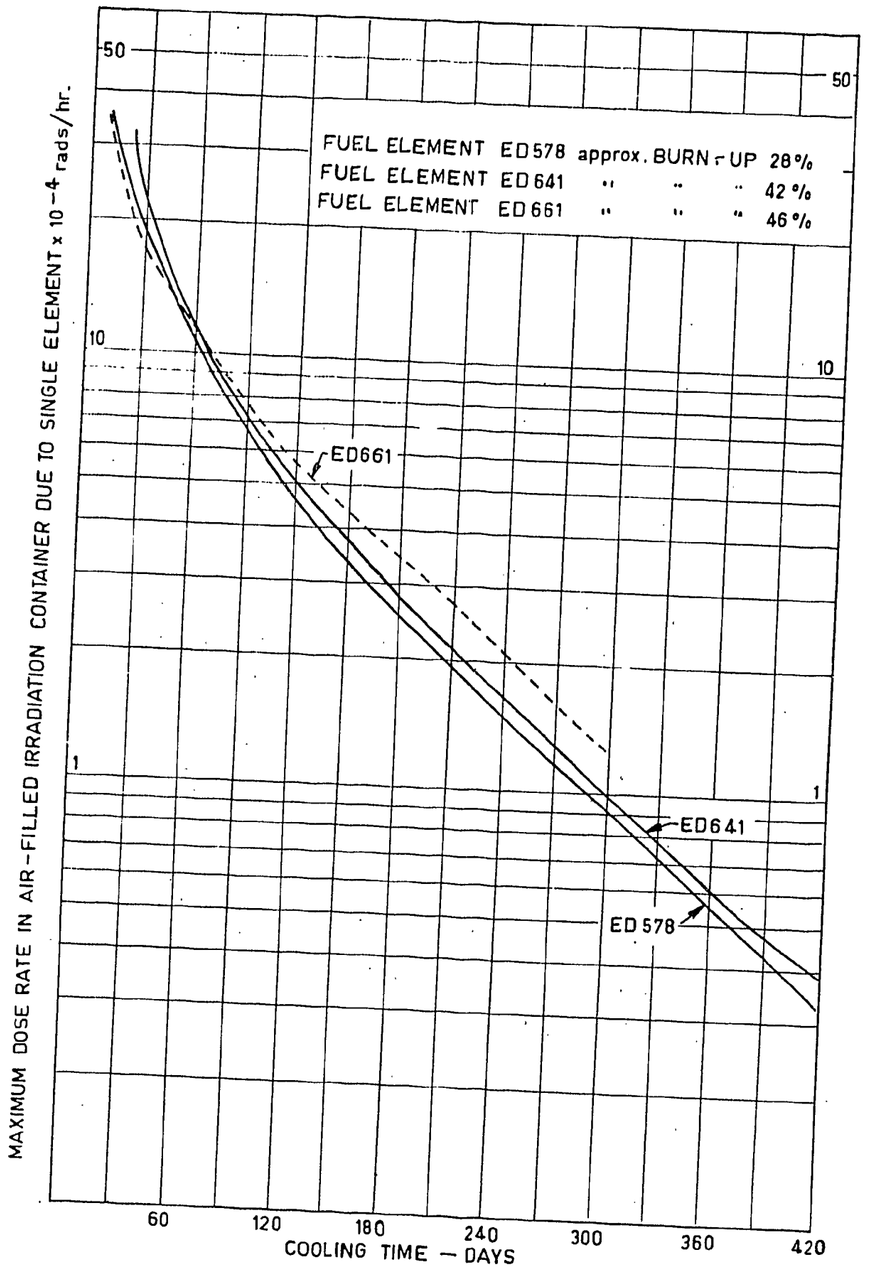



**FIGURE 4. DECAY GRAPHS FOR SINGLE SPENT FUEL ELEMENTS OVER EXTENDED COOLING TIME**

P1001

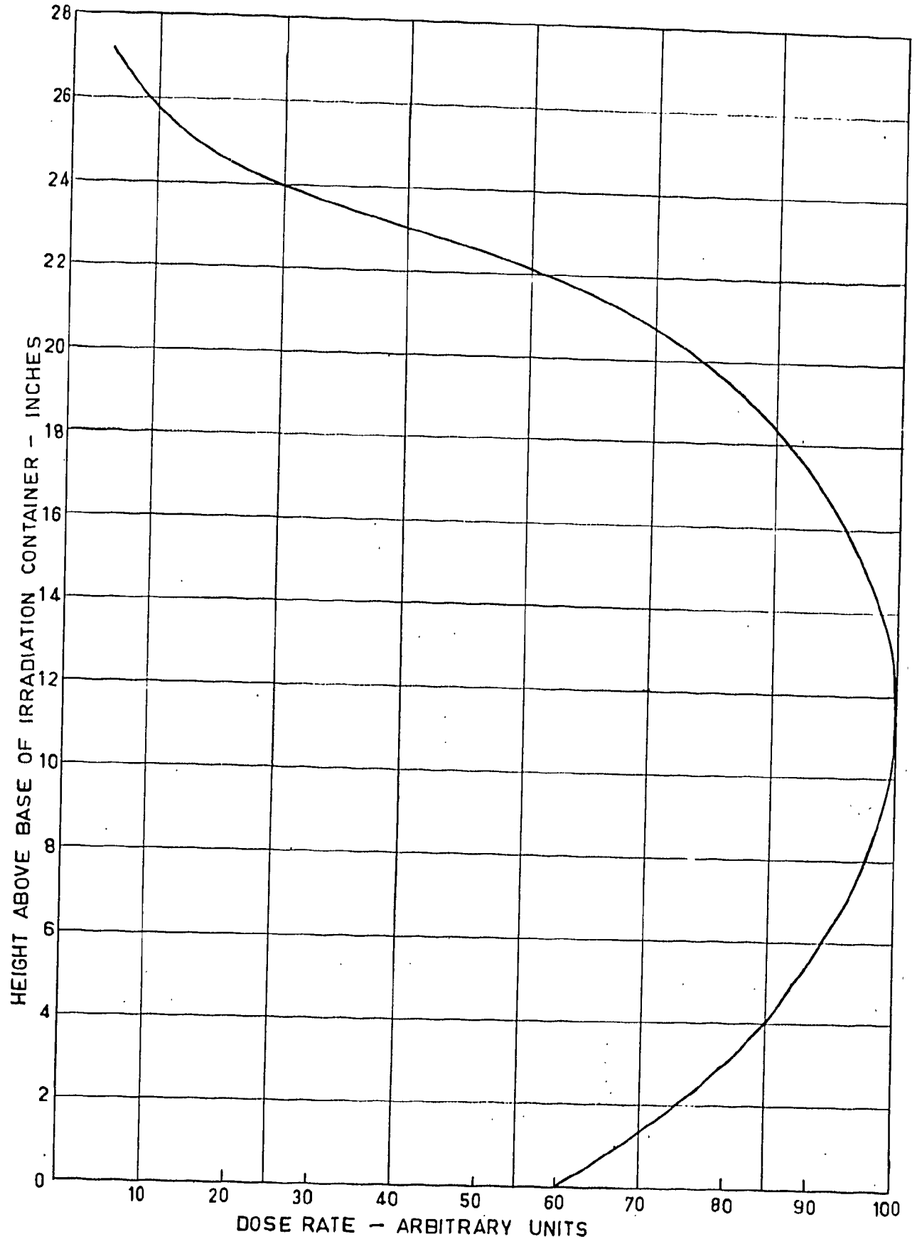



**FIGURE 5. TYPICAL DOSE RATE DISTRIBUTION ALONG THE CENTRAL AXIS OF A 1.5 INCH DIAMETER AIR-FILLED IRRADIATION CONTAINER IN A SPENT HOLLOW FUEL ELEMENT**

P1001