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EPITHERMAL BEAM DEVELOPMENT AT THE BMRR: DOSIMETRIC EVALUATION

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INTRODUCTION

The utilization of an epithermal neutron beam for neutron capture therapy (NCT) is desirable because of the increased tissue penetration relative to a thermal neutron beam. Over the past few years, modifications have been and continue to be made at the Brookhaven Medical Research Reactor (BMRR) by changing its filter components to produce an optimal epithermal beam. An optimal epithermal beam should contain a low fast neutron contamination and no thermal neutrons in the incident beam. Recently a new moderator for the epithermal beam has been installed at the epithermal port of the BMRR and has accomplished this task.(1) This new moderator is a combination of alumina (Al_2O_3) bricks and aluminum (Al) plates. A 0.51 mm thick cadmium (Cd) sheet has reduced the thermal neutron intensity drastically. Furthermore, an 11.5 cm thick bismuth (Bi) plate installed at the port surface has reduced the gamma dose component to negligible levels. In order to compare various filter configurations for best optimization,(2) the following parameters have been measured on the beam axis, directly in front of the epithermal port:

1) thermal neutron fluence rate free in air
2) epithermal neutron fluence rate free in air
3) fast neutron fluence rate free in air
4) thermal neutron fluence rate in a polyethylene cylindrical head phantom as a function of distance along the axis of the phantom.
5) fast neutron dose rate in soft tissue in air
6) gamma dose rate in soft tissue in air

Foil activation techniques have been employed by using bare gold and cadmium-covered gold foil to determine thermal as well as epithermal neutron fluence. Fast neutron fluence has been determined by indium foil counting. Fast neutron and gamma dose in soft tissue, free in air, is being determined by the paired ionization chamber technique, using tissue equivalent (TE) and graphite chambers. Thermoluminescent dosimeters (TLD-700) have also been used to determine the gamma dose independently.

This paper describes the methods involved in the measurements of the above mentioned parameters. Formulations have been developed and the various corrections involved have been detailed.

DISCLAIMER

FOIL ACTIVATION ANALYSIS

It occurs quite frequently that transmutations induced by neutrons have radioactive species as product nuclides. The activity of the product radioisotope can often be used to measure the amount and energy distribution of the neutron flux which induced the radioactivity. A detector employing this principle is referred to as a neutron-activation detector.

The thermal, epithermal and fast neutron fluence rate at the BMRR epithermal port facility were determined using thin gold, copper, and indium foils. Here, thermal neutrons (energy range from 0 to 0.4 eV) refers not only to the maxwellian component of the spectrum, but also to the much weaker slowing down component, ideally proportional to 1/E. Epithermal neutrons have been defined as having neutron energies between 0.4 eV to 10 keV and fast neutrons from 10 keV to the end-point of the fission spectrum.

The thermal neutron fluence rate, ϕ_{th}, has been determined via measurements of the induced gamma activity in gold and copper foils (gold foils of thickness 0.0127 mm, average mass 9 mg and average diameter 6.8 mm; copper foils of average mass 5 mg with the same dimensions as gold). Gold has a resonance at an energy of 4.9 eV, the effect of this resonance in determining the thermal neutron fluence ϕ_{th} was eliminated by the cadmium difference method. A Cadmium cover of thickness 0.76 mm was used to sandwich the gold foil, which was then irradiated at the reactor's epithermal port surface. The induced activity of the gold foil was measured with a NaI (Tl) well type detector (having an absolute efficiency of 25%). The counting system was calibrated by absolute β and γ counting as well as with the BNL's sigma-pile standard neutron source. The thermal neutron fluence rate (ϕ_{th}) was calculated using the following equation:

$$\phi_{th} = \phi_{bare} \cdot F_{Cd}\, \phi_{Cd} \qquad (1)$$

where,

ϕ_{bare} is the fluence rate measured with a bare gold foil
ϕ_{Cd} is the fluence rate measured with a Cd-covered gold foil, and
F_{Cd} is the correction factor that takes into account the absorption of resonance neutrons by cadmium.

The correction factor, F_{Cd}, depends on a number of parameters such as the thickness of the Cd cover, the thickness of the gold foil, and the angular distribution of the resonance neutrons, etc. For a 0.76 mm Cd cover, used for a 9 mg gold foil, the correction factor used is 1.02.[3,4]

The ratio of the activity of a bare gold foil to that of the Cd-covered gold foil is defined as the cadmium ratio (CdR), and this parameter is a measure of the degree of thermalization in the beam. The activity of bare and Cd-covered foils are measured at the same point on the port face. Ideally, the gold foil detector should be so thin that the flux would not be perturbed, but in practice, the foils used are always thick enough to produce

self-shielding. The method used to correct the flux depression and the self-shielding gives,[3]

$$CdR(0) = CdR(X) \cdot Q(X) + F_{Cd} (1 - Q(X)) \qquad (2)$$

where the parameter Q(X) is equal to 0.53 for a 25 mg/cm^2 thick gold foil, and CdR(0) is the cadmium ratio of the detector (gold foil) corrected for the flux depression and the self-shielding effects.

The measured cadmium ratio has been utilized in the determination of the epithermal neutron fluence rate. The ratio of the thermal flux to the resonance or epithermal flux in terms of the effect on the particular detector is given by[5]

$$\frac{\text{Thermal flux}}{\text{Resonance flux}} = CdR - 1 \qquad (3)$$

The epithermal resonance of the gold foil at 4.9 eV has been well determined and the resonance integral has a value of 1558 barns. By using thermal activation cross section of 98.8 barns, the epithermal neutron fluence per energy decade was measured.

The fast neutron fluence was measured in accordance with the In(n,n') reaction. Indium foils of thickness 0.127 mm with a mass of 98 mg/cm^2 were shielded by an enriched ^{10}B sphere to eliminate the 1.45 eV indium resonance. Counting was done using a High Purity Ge detector whose efficiency was determined by a standard ^{152}Eu source. Table I includes the various components of the measured neutron fluence rates at 1 MW power at the epithermal port of the BMRR.

IONIZATION CHAMBERS

A prerequisite for biomedical studies is that energy dissipation in irradiated material be determined with a sufficient degree of accuracy and precision. This is a difficult task considering the complexity of dose determination due to inhomogeneities in tissue composition and density. In addition, specification of the absorbed dose, however, does not account for the microscopic distribution of energy deposition, i.e., the radiation quality. However, this microscopic distribution has to be considered since the degree of damage depends not only on the total amount of energy deposited but also on the spatial distribution of the energy deposition, i.e., on LET. Because of the differences in relative biological effectiveness of the various radiation components, it is necessary to determine the neutron absorbed dose from fast neutrons, D_N, the gamma absorbed dose, D_γ, and the absorbed dose from the $^{14}N(n,p)^{14}C$ reaction, D_p, in the mixed field separately.

Table I. Measured parameters at the BMRR (at 1 MW power) for the $Al_2\mathcal{C}_3$/Al filtered Epithermal Beam

i)	Epithermal neutron fluence rate from 0.4 eV to 10 keV	6×10^8 neutrons/cm^2- sec
ii)	Thermal neutron fluence rate up to 0.4 eV	2.4×10^7 neutrons/cm^2- sec
iii)	Fast neutron fluence rate for energy >1.2 MeV	3.2×10^7 neutrons/cm^2- sec
iv)	Cadmium Ratio	1.05
v)	Fast neutron dose rate for soft tissue in air using TE ionization chambers	1.75 rads/min
vi)	Gamma dose rate for soft tissue in air using graphite ionization chambers and TLDs	0.4 rads/min
vii)	Peak thermal neutron fluence rate generated inside a cylindrical head phantom filled with tissue equivalent liquid	8.5×10^8 neutrons/cm^2-sec
viii)	Relative biological effectiveness (RBE) obtained with V-79 Chinese hamster cells	1.8

The use of a calibrated A-150 plastic tissue-equivalent (TE) ionization chamber filled with TE gas is generally considered to be the most satisfactory method for determining the total absorbed dose in mixed neutron/gamma fields for biomedical applications. Ionization chambers respond to both neutrons and gammas. Thus, one of the principal problems in neutron dosimetry with chambers is the separation of the absorbed dose contributed byeach of these radiations. This separation was accomplished by using a tissue-equivalent chamber with tissue equivalent gas (methane-based) to measure the total absorbed dose and by using a neutron insensitive graphite wall chamber filled with CO_2 gas to essentially measure the gamma component. Finally, by subtracting the gamma component from the total absorbed dose, the neutron absorbed dose can be determined.[6]

The fast neutron and gamma dose to tissue in air for the epithermal beam at the port surface of the BMRR was determined using Far West Technology ionization chambers. Charge was measured using a Keithley 614 electrometer. Two different size chambers were used: a) both, a tissue equivalent chamber and, a graphite chamber with a volume of 0.1 cc and b) a tissue equivalent chamber with a volume of 1 cc and a graphite chamber with a volume of 2.5 cc. The tissue equivalent methane-based gas and CO_2 gas were obtained from Methason gas products. During the course of the measurements, the 25 x 25 cm^2 port was shielded by a ^6Li metal plate to reduce the thermal neutron component. All chambers were calibrated by both Far West Technology and the calibration facility of Brookhaven National Laboratory (BNL); and there was an excellent agreement between the calibrations. Calibration was performed at a fixed pressure and temperature, thus necessitating a correction for these two parameters for different pressure and temperature conditions, which will be discussed later along with other correction factors.

The fundamental equation for the determination of the absorbed dose rate $\dot{D}_m$, in the wall material adjacent to the cavity of the chamber is[7]

$$\dot{D}_m = \frac{\dot{Q}}{M} \cdot \frac{\bar{w}}{e} \cdot s_{m,g} \qquad (4)$$

where $\dot{Q}$ is the ionization current
M is the mass of the cavity gas
$\bar{w}$ is the average energy expended to produce an ion pair
e is the fundamental electronic charge, and
$s_{m,g}$ is the ratio of average mass stopping power of the wall of the chamber (A-150 plastic) to the gas.

The absorbed dose rate $\dot{D}_m$ in the unknown field relative to the absorbed dose rate, $\dot{D}^c_m$, in the calibration field is

$$\frac{\dot{D}_m}{\dot{D}^c_m} = \frac{\dot{Q}}{\dot{Q}_c} \cdot \frac{M_c}{M} \cdot \frac{\bar{w}}{\bar{w}_c} \cdot \frac{s_{m,g}}{s^c_{m,g}} \qquad (5)$$

Since the chamber volume and gas composition are constant, the mass ratio can be replaced by pressure and temperature. At a point in the mixed (n,γ) field, where $\dot{D}^n_m$ and $\dot{D}^\gamma_m$ are the absorbed dose rates in tissue from neutrons and gammas respectively, we get.

$$\frac{\dot{Q}}{\dot{Q}_c}\frac{M_c}{M} = \frac{\bar{w}^c \; s^c_{m,g} \; \dot{D}^n_m}{\bar{w}^n \; s^n_{m,g} \; \dot{D}^c_m} + \frac{\bar{w}^c \; s^c_{m,g} \; \dot{D}^\gamma_m}{\bar{w}^\gamma \; s^\gamma_{m,g} \; \dot{D}^c_m} \qquad (6)$$

where $\bar{w}^n$ and $\bar{w}^\gamma$ are the average energy expended to produce an ion pair by the neutron and the gamma components of the mixed field.[8] $s^n_{m,g}$ and $s^c_{m,g}$ are the ratio of the average stopping power of each of the components to the wall material and the gas.[9]

The calibration fields of the ^{60}Co and ^{137}Cs sources at BNL are calibrated in units of the exposure rate, $\dot{X}^c$, free in air. We can obtain the absorbed dose rate $\dot{D}^c_m$ in the wall of the chamber from the exposure rate $\dot{X}^c$, by using the following relation:

$$\dot{D}^c = f \cdot (\overline{\mu/\rho})^{m,air} \cdot \lambda \cdot \dot{X}^c \qquad (7)$$

$$\dot{D}_m^c = f \cdot \left(\overline{\mu/\rho}\right)_{energy}^{m,air} \cdot \lambda \cdot \dot{X}^c \qquad (7)$$

where

f is a conversion factor from Roentgen to rads and is equal to 0.873 rads/R.

$\left(\overline{\mu/\rho}\right)_{energy}^{m,air}$ is the average ratio of the mass energy absorption coefficient of the gamma field in the wall and in the air of calibration facility.[10]

λ is a factor that takes into account the attenuation of the calibration field by the wall of the chamber and

$\dot{X}^c$ is the exposure rate free in air of the calibration field.

Now, by combining equations (6) and (7), we obtain:

$$\dot{D}_m^n = 0.873 \cdot \left(\overline{\frac{\mu}{\rho}}\right)_{energy}^{m,air} \cdot \lambda \cdot \dot{X}^c \cdot \frac{\dot{Q}}{\dot{Q}^c} \cdot \frac{M_c}{M} \cdot \frac{\bar{w}^c \, s_{m,g}^{\gamma} \, \dot{D}_m^{\gamma}}{\bar{w}_{m,g}^{\gamma} \, s^{\gamma} \, \dot{D}_m^c} \cdot \frac{\bar{w}^n \, s_{m,g}^n}{\bar{w}^c \, s_{m,g}^c} \qquad (8)$$

where $\dot{Q}$ and $\dot{Q}^c$ are the measured values of currents in the two fields.

Now, the entrance absorbed dose rate to the tissue, free in air, due to the fast neutron component of the mixed field $\dot{D}_{s.t.}^n$ can be calculated as shown below:

$$\dot{D}_{s.t.}^n = \lambda \cdot \frac{k_{s.t.}^n(E)}{k_m^n(E)} \cdot \dot{D}_m^n \qquad (9)$$

Here, $k_{s.t.}^n(E)/k_m^n(E)$ is the ratio of the kerma in soft tissue and the wall material over the neutron spectrum.

The quantity $\bar{w}_c \cdot s_{m,g}^c \cdot \dot{D}_m^{\gamma}/\bar{w}^{\gamma} \cdot s_{m,g}^{\gamma} \cdot \dot{D}_m^c$ is usually written as h_T and defined as the sensitivity of the chamber to the mixed field gamma rays relative to the sensitivity of the radiation used for the calibration. A graphite chamber filled with CO_2 gas has a lower sensitivity for neutrons and thus was used to determine the gamma component of the mixed field.

Finally, by substituting the values of the various parameters in Equation (6) for TE and Graphite chambers, we obtain

$$\dot{Q}^{n,\gamma} = 1.035\ \dot{D}_m^{\gamma} + 0.92\ \dot{D}_m^{n} \qquad \text{for TE chamber} \qquad (10)$$

$$\dot{Q}^{n,\gamma} = 1.036\ \dot{D}_m^{\gamma} + 0.088\ \dot{D}_m^{n} \qquad \text{for Graphite chamber} \qquad (11)$$

Here, the kerma values over the neutron spectrum, were taken from the ICRU report 26. $\dot{Q}^{n,\gamma}$ is the total registered current converted to dose rate using the calibration constant of the chambers. These two coupled equations have been solved to give the fast neutron and gamma dose component of the mixed field radiation.

Although ionization chambers are the best devices for measuring the fast neutron and gamma components of the mixed field, results obtained by these measurements are susceptible to large errors. As has been pointed out earlier, dose determination requires accurate values of w and kermas. Calculation of fast neutron dose also requires a precise knowledge of the neutron spectrum in order to avoid large errors. Other errors can be minimized by appropriately applying a correction factor.

Ionization chamber results are determined by measuring the ionizations produced in the cavity by charged particles created in the wall, central electrode and gas. The relative contributions from the wall material and gas are dependent on the neutron energy spectrum. The wall of a chamber must be thick enough to establish secondary charged particle equilibrium. The minimum thickness is determined by the maximum range of the secondaries. However, walls also causes attenuation of the primary radiation. Under conditions of charged particle equilibrium, absorbed dose is equal to kerma. A 5 mm thick TE plastic cup reduces the response by 10%. For the determination of kerma free-in-air, the readings made from wall thicknesses in excess of the minimum value require for the establishment of charged particle equilibrium are generally extrapolated to zero wall thickness and so in this case a correction factor has been applied.[11]

The calibration factors supplied with the Far West Technology ionization chambers are referenced to 760 mm Hg at 22°C. Also, calibration at the Brookhaven calibration facility was performed under differing pressure and temperature conditions than when the chambers were used to measure dose in the mixed field radiation; therefore, a correction was applied as follows:

$$\text{Factor } F = \frac{273 + T}{273 + t} \cdot \frac{P}{p} \qquad (12)$$

where T and P are the temperature and pressure at the time of calibration, and t and p are true respective values at the time of the mixed field measurement.

Other factors which have been incorporated in the final evaluation of the dose to the soft tissue in air are:

i) saturation effects
ii) polarity effects
iii) gas flow dependence
iv) offset and leakage current
v) influence of ^{6}Li metal cover
vi) stem scattering

The fast neutron dose and gamma dose for soft tissue in air for the epithemal beam at the BMRR have been measured using FWT ionization chambers and results, after incorporating the appropriate correction factors, are shown in Table I.

THERMOLUMINESCENT DOSIMETERS

Thermoluminescent devices have been used extensively for the dosimetry of X-, γ-, and β-rays. In neutron dosimetry, the main use of thermoluminescent dosimeters (TLD) has been for the measurement of the associated gamma rays. Thermoluminescence (TL) is a phenomenon exhibited by a variety of organic and inorganic materials. The latent luminescence induced by the ionizing radiation (basically by trapped electrons in host material) is released at high temperature. The proportionality between the released thermoluminescent light and the absorbed dose is the basis for this dosimetric technique.

The incident gamma ray component of the total dose rate in soft tissue in air at the epithermal port of the BMRR was determined by using LiF-TLD rods (TLD-700), obtained from Harshaw Chemical Corp., of dimensions 1 x 1 x 6 mm^3. During reactor irradiations the TLD dosimeters were placed in a plastic bottle containing ^{6}Li-enriched (~95.6% ^{6}Li) LiF powder and were positioned on the central axis of a cylindrical teflon base (14 mm x 7.2 mm diameter). In all cases the plastic bottle was placed on its horizontal axis of symmetry, thus assuring that the minimum thickness of shielding for the TLD was 0.5 g/cm^2 of ^{6}LiF, which is equivalent to 2.1 mm thickness of ^{6}Li metal. In order to verify that the TLDs were well protected from the thermal neutrons, gold wires were inserted between the teflon base and the plastic inner bottle. The gamma activity of the gold wires corresponded to 10^6 thermal neutrons/cm^2-sec at 1 MW. It has been determined for this batch of TLDs that a fluence of 10^{10} thermal neutron/cm^2-sec at 1 MW would induce a signal equivalent to $\simeq$1.2 rads in soft tissue. Therefore, it was concluded that the TLD rods were adequately protected from the themal neutron beam, and that they produced negligible signal. In a similar fashion, it was assumed that the contribution to the TL signal from high energy neutrons was insignificant, due to their inability to induce a TL response. (This assumption is under question, due to the lack of

adequate knowledge of the neutron spectrum.)

Standard heat treatment was applied to the TLD-700. The one-hour pre-irradiation annealing at 400°C in a stainless steel base was followed by a slow cooling rate to room temperature (15 min furnace-cool, followed by a 30 min air-cool). The post-irradiation annealing (100°C in air for 15 min) in a teflon base, was followed with a 45 min air cooling. This post-irradiation treatment helps in eliminating peaks 2 and 3 from the spectrum of the glow curve of the Lif-700 dosimeters. The emitted TL signal (integrated between room temperature and 350°C) was detected using Harshaw's model 2000 A and B and also on Harshow's model 4000 thermoluminescence reader in 30 seconds. The glow curve of each dosimeter was recorded on paper as well as on computer mass media storage. The high temperature limit of 350°C was selected to reduce the black body radiation contribution. There are several problems which complicate the interpretation of the glow curve results. For example, after irradiation with neutron or gammas the glow curves sometimes have different shapes. This is one of the reasons that the entire glow curve was integrated to assure proper interpretation of the results.

The gamma dose rate component $\dot{D}_{\gamma}^{s.t.}$ in an elemental mass of soft tissue, free in air, has been determined using the following relation:

$$\dot{D}_{\gamma}^{s.t.} = (\mu/\rho)_{energy}^{s.t./tef.} \cdot \lambda \cdot C \cdot \dot{D}_{Co}^{LiF} \qquad (13)$$

where $(\overline{\mu/e})_{energy}^{s.t./tef.}$ is the ratio of mass energy absorption coefficients in soft tissue and teflon, λ is the linear attenuation coefficient for gamma rays in teflon, C is the cavity theory correction for gamma rays and $\dot{D}_{Co}^{LiF}$ is the dose rate in the TLDs located in the teflon base and irradiated with standard calibrated ^{60}Co and ^{137}Cs sources. Teflon helps in producing charged particle equilibrium and provides a base for placement of TLD rods on the reactor port. Teflon was used rather than other hydrogenous materials as it does not produce any contaminating radiation. Calculations were made assuming that the gamma component from the reactor has an energy of 2.2 MeV. At present we have no information on the gamma spectrum.

At least 8 dosimeters were used to determine the background (electronic noise of the instrument, black body radiation, etc.) components during each run. Cobalt-60 and cesium-137 sources of the BNL calibration facility were used to calibrate TLDs for each run. The same teflon block and cylinders were used throughout the calibration. Care was exercised in converting the exposure rate free in air to the dose rate in the dosimeter material that was placed in the teflon block. The gamma dose rate in soft tissue, free in air, was determined by comparing the total light output to the light output of the calibrated TLDs light output and converting the dose rate to soft tissue.

The absorbed dose from the $^{14}N(n,p)^{14}C$ reaction has been calculated as follows:

$$^{14}N_D = 1.602 \times 10^{-8} \; EFN\sigma\Phi \qquad (14)$$

where $^{14}N_D$ is the absorbed dose in rads due to nitrogen

1.602×10^{-8} is the conversion factor, MeV to 100 ergs

E is the energy released, in MeV per event

F is the fraction by weight of the nitrogen in tissue

N is the number of atoms per gram of tissue of the nitrogen

σ is microscopic absorption cross section $\frac{cm^2 \cdot event}{atom}$

and Φ is the thermal neutron fluence $\frac{neutrons}{cm^2}$

For the $^{14}N(n,p)^{14}C$ reaction the energy released per event E is 0.585 MeV, and the microscopic absorption cross section σ is 1.85×10^{-24} cm^2. Using nitrogen content in brain tissue by weight fraction as 1.84%, the nitrogen or thermal neutron dose has been calculated using the following equation:

$$^{14}N_D = 0.745 \times 10^{-9} \; F\Phi \text{ in rads}$$

In a similar fashion, the $^{10}B(n,\alpha)^7Li$ dose has been calculated at various depths in the head phantom by measuring the thermal neutron fluence rate by gold and copper foils. The ^{10}B dose to the tissue is, (assuming uniform distribution of boron over the tissue volume)

$$^{10}B_D = 1.602 \times 10^{-8} \; EFN\sigma\Phi$$

for the ^{10}B reaction, the energy released per event is 2.34 MeV and the microscopic absorption cross section, σ, used in calculating the dose has the value 3838×10^{-24} cm^2. Finally the equation used to calculate the dose is:

$$^{10}B_D = 8.66 \times 10^{-6} \; F\Phi \text{ in rads}$$

V-79 Chinese hamster cells were irradiated in suspension as detailed elsewhere.[12] Neutron irradiations were carried out at the epithermal

port of the BMRR at a distance of 1.2 cm from the port face. X-irradiation was carried put at the 250 kVp Maxitron, 60 cm from the source, using 0.5 mm copper and 1.0 mm aluminum filters. Following the irradiation, cells were plated for survival assay. The ratio of the D_0 values obtained from the linear portion of each survival curve demonstrated a RBE of ~1.8.

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