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FERROUS SULPHATE (FRICKE) DOSIMETRY  
IN A FAST NEUTRON AND A  $^{60}\text{Co}$   
RADIOTHERAPEUTICAL BEAMS

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**WYDANO NAKŁADEM  
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NA PRAWACH RĘKOPISU**

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## Abstract

Ferrous sulphate (Fricke) dosimetry was performed in the fast neutron beam produced at the U-120 cyclotron of the Institute of Nuclear Physics, and in a  $^{60}\text{Co}$  gamma-ray therapeutical beam at the Centre of Oncology in Kraków. The G-value was measured for the neutron component in the mixed (neutron + gamma) field of the fast neutron beam, where the mean neutron energy is 5.6 MeV. This value is  $G_n = 8.1 \pm 0.8$ . Track structure theory calculations were made using energy spectra of charged secondary particles generated in water by the MRC Hammersmith fast neutron beam (mean neutron energy 7.6 MeV, measured value of  $G_n = 9.4 \pm 0.6$ ), yielding the calculated value of  $G_n = 8.61$ . Fricke dosimetry of the  $^{60}\text{Co}$  beam indicates that the absolute value of gamma-ray dose at the Centre of Oncology may be underestimated by about 3%.

# DOZYMETRIA FRICKEGO DLA RADIOTERAPEUTYCZNEJ WIĄZKI NEUTRONÓW

## PRĘDKICH I PROMIENI GAMMA

### Streszczenie

Za pomocą dozymetru siarczkowo-żelazowego (Frickego) zmierzono dawki w terapeutycznej wiązce neutronów prędkich produkowanych w cyklotronie U-120 IFJ, oraz w wiązce promieni gamma od źródła  $^{60}\text{Co}$  stosowanej do radioterapii w Centrum Onkologii w Krakowie. Zmierzono wartość liczby G dla składowej neutronowej mieszanego pola (neutron + gamma) wiązki neutronów prędkich, gdzie średnia energia neutronów wynosi 5.6 MeV. Wynosi ona  $G_n = 8.1 \pm 0.6$ . W oparciu o model struktury śladu wykonano obliczenia teoretyczne, w których wykorzystano znane z innych prac widma naładowanych cząstek wtórnych generowanych w wodzie przez neutrony prędkie z cyklotronu MRC Hammersmith (średnia energia neutronów 7.6 MeV, zmierzona wartość  $G_n = 9.4 \pm 0.6$ ), otrzymując obliczoną wartość  $G_n = 8.61$ . Pomiar dozymetrem Frickego wykonany dla wiązki  $^{60}\text{Co}$  wykazuje, że wartość dawki bezwzględnej promieniowania gamma w Centrum Onkologii w Krakowie może być zaniżona o ok. 3%.

# ФРИКЕ-ДОЗИМЕТРИЯ ДЛЯ РАДИОТЕРАПЕВТИЧЕСКОГО ПУЧКА БЫСТРЫХ НЕЙТРОНОВ И ГАММА-ЛУЧЕЙ

## Содержание

Используя сульфатно-железистый (Фрикe) дозиметр определены дозы в терапевтическом пучке быстрых нейтронов, генерированных циклотроном Y-120 Института Ядерной Физики в Кракове, а также в пучке гамма-лучей источника  $^{60}\text{Co}$  используемого в радиотерапии в Онкологическом Центре в Кракове. Определено число G-фактора для нейтронной составляющей смешанного поля (нейтрон+гамма) пучка быстрых нейтронов, где средняя энергия нейтронов - 5.6 MeV,  $G_n = 9.1 \pm 0.6$ . Опираясь на модель структуры трека произведен теоретический расчет, в котором использованы взятые из других работ спектры заряженных вторичных частиц, генерированных в воде быстрыми нейтронами циклотрона MRC Hammerwith (средняя энергия нейтронов - 7.6 MeV, экспериментальное значение  $G_n = 9.4 \pm 0.6$ ), получая теоретическое значение числа  $G_n = 8.61$ . Фрикe-дозиметрия на пучке  $^{60}\text{Co}$  показывает, что величина абсолютной дозы гамма-излучения в Онкологическом Центре может быть занижена приблизительно на 3%.

## 1. Introduction

Of the chemical systems available for dosimetry, the ferrous sulphate (Fricke) dosimeter is recognised to be the most suitable with respect to accuracy, reproducibility and linearity (1). Considerable attention has been devoted to standardizing the preparation of this system (2) and to measuring its response after doses of gamma rays (principally  $^{60}\text{Co}$ , (3)), after a variety of heavy charged particle irradiations (4), as well as after doses of fast neutrons of various energies in the range 0.1 to 18 MeV (5,6).

Only calorimetric devices and the Fricke dosimeter can currently be used for absolute dosimetry. Such dosimetry is of obvious importance to radiation therapy facilities, including those using cyclotron-produced beams of fast neutrons. The neutron beam in Krakow (7), of mean energy 5.6 MeV, in many ways resembles that of the MRC in Hammersmith (mean energy 7.6 MeV) where several accurate measurements using the Fricke dosimeter have been performed (8).

Following a recent improvement of the formula describing the radial distribution of dose around the path of a heavy ion (9), the measured response of the Fricke dosimeter to charged particle irradiations has been adequately described (10) in terms of 1-hit detector properties, according to Katz's track structure theory (11,12). It is then possible to calculate the response of the Fricke dosimeter to a beam of fast neutrons using this theory.

Up to now, no Fricke dosimetry of the fast neutron beam has been performed in Krakow. The principal aim of this work was therefore to develop an acceptable ferrous sulphate system and perform beam dosimetry in order to determine the response of the Fricke dosimeter to our fast neutron beam, as related to its response to a  $^{60}\text{Co}$  radiotherapy beam used at the Centre of Oncology in Krakow. The latter measurement would also enable an absolute determination of the Krakow gamma-ray dosimetry, against routinely used ionization chambers (13,14) and against the new alanine dosimeter which has recently been tested in Krakow in a joint Danish-Polish effort (15).

We also made theoretical calculations in order to compare the results obtained from our measurements with those calculated. In our track structure calculations, due to the lack of input data pertaining to the energy spectrum of the neutron beam in Krakow, we had to use the spectra of charged secondaries following irradiation of water by fast neutrons of the Hammersmith beam, known from other work (11). The results of our calculation could then test the sensitivity of the model to the differences between the energy spectra of the neutron beams at Krakow and at Hammersmith.

## 2. Materials and methods

### 2.1 The Fricke dosimeter

The preparation of the dosimeter solution and its subsequent measurements after irradiations were carried out strictly according to the Standard Test Method (ASTM Designation D 1671-72) (2), with minor modifications in the recommended spectrophotometer calibration procedure.

#### 2.1.1 Standard recommendations and formulae

The method of preparing the solution of ferrous ammonium sulphate (0.001 M in  $\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2$ , 0.001 M in NaCl, and 0.8 N in  $\text{H}_2\text{SO}_4$ ), and its subsequent use in dosimetry, are described in detail by the ASTM Standard (2).

As it is well known, the change of absorbance of the dosimetric solution caused by chemoradiative oxidation of ferrous to ferric ions is proportional to the dose absorbed. Changes in absorbance are measured at the UV wavelength of 304 nm, against the absorbance of the unexposed dosimetric solution. The concentration of  $\text{Fe}^{2+}$  ions, hence the absolute dose, can then be determined directly.

The absorbed dose, D (in cGy), is calculated from the following formula (2):

$$D = (9.65 \times 10^6 \times f \times (OD_e - OD_u)) / (E \times G \times \rho) \quad (1)$$

where:

- OD<sub>e</sub> = absorbance of the exposed solution,
- OD<sub>u</sub> = absorbance of the unexposed solution,
- f = dilution factor, final volume/initial volume (when needed),
- G = the number of molecules reacting per 100 electron volts absorbed. (G = 15.6),
- E = molar extinction coefficient
- ρ = density of undiluted solution (1.02 g/ml)

As the molar coefficient of extinction depends on temperature, the following temperature correction to the measured dose is recommended (2):

$$D(T_1) = D(T_2) [1 + 0.007 \times (T_1 - T_2)]^2 \quad (2)$$

the formula in (2), p. 429, is erroneous



where:

- $T_1$  = temperature at which the calibration curve was prepared, deg C, and  
 $T_2$  = temperature at which the unknown sample is read, deg C,  
 $D(T_1)$  = dose (cGy) measured at temperature  $T_1$ , and  
 $D(T_2)$  = dose (cGy) referred to temperature  $T_2$ .

### 2.1.2 Reagents and ampoules

The dosimetric solution containing 1 mM ferric ammonium sulphate and 1 mM sodium chloride in 1 l of 0.8 N sulphuric acid was prepared using re-distilled water and analytic-grade reagents. A fresh solution was prepared before each exposure run and samples, each containing 3 ml of solution, were placed in ampoules made of "Termisil" glass (first batch) or of "Sylvit" glass (second batch). Ampoules were flame-sealed. Before placing the dosimetric solution in them, ampoules were filled with a solution of ferrous ammonium sulphate, irradiated with 250 kVp X-rays to a dose of c. 500 Gy and emptied, in accordance with ASTM recommendations (2). The dosimetric samples were irradiated in the same ampoules the internal diameter of which was 17.2 mm and wall thickness 1.1 mm.

### 2.1.3. Spectrophotometry.

All optical density measurements were performed using a VSU 2-P spectrophotometer (Carl Zeiss, Jena, GDR) at analytical wavelength of 305 nm, 0.6 mm slit width, and using quartz cuvettes of 10 mm thickness. The absorbance readouts were temperature corrected whenever necessary using the temperature coefficient of +0.7% per deg C.

Solutions used to determine the spectrophotometry calibration curve were prepared as follows: an exact aliquot of 4.8198 g  $\text{Fe NH}_4(\text{SO}_4)_2 \cdot 12 \text{H}_2\text{O}$  was diluted in 1 l 0.8 N sulphuric acid (solution A); 10 ml of this solution was diluted with 0.8 N sulphuric acid to a volume of 100 ml (solution B). Next, 1, 5, 10, 15, 20 and 25 ml portions of solution B were transferred to 100 ml volumetric flasks and filled with 0.8 N sulphuric acid up to the 100 ml marker. Absorbances of thus diluted solutions were then read using the spectrophotometer, with the result given above.

The molar coefficient of absorption was found to be 2106  $\text{l mol}^{-1} \text{cm}^{-1}$  at a temperature of 18.5 °C. (In the range of calibration concentrations,  $C = (1.8 - 25) \times 10^{-5} \text{ mol l}^{-1}$  of  $\text{Fe}^{3+}$  ions, the calibration curve was fitted by linear regression as  $A (\text{cm}^{-1}) = -4.01 \times 10^{-2} (\text{cm}^{-1}) + 2106 (1 \text{ mol}^{-1} \text{cm}^{-1}) \times C (\text{mol l}^{-1})$ , with a coefficient of determination  $R^2 = 0.99941$ ). When corrected to 24.5°C, the value of this coefficient of absorption is 2194  $\text{l mol}^{-1} \text{cm}^{-1}$ .

$\text{mol}^{-1}\text{cm}^{-1}$ , well within the recommended range  $2195 \pm 10$   $\text{l mol}^{-1}\text{cm}^{-1}$  (2).

No temperature correction was made in the calculations of doses to account for the variations of temperature at which samples were irradiated as these never exceeded the range 18-24°C.

## 2.2. Radiation sources and dosimetry.

The gamma-ray irradiations were carried out with one of the  $^{60}\text{Co}$  sources at the Centre of Oncology in Krakow, a SIEMENS Gammatron S radiotherapy unit, of present activity of c.  $8 \times 10^{13}$  Bq. Fricke dosimeter ampoules were exposed at a dose rate of c.  $4 \times 10^{-2}$  Gy/s, under the control of a Baldwin-Farmer air ionization chamber of  $0.6 \text{ cm}^3$  connected to an IONEX type 2500 exposure meter. Ampoules were placed in a Perspex holder of 5 mm thickness to ensure electron equilibrium, and the absolute accuracy of exposure assessment was c. 2 %. In two separate runs seven exposures were made, ranging from about 50 Gy to 200 Gy.

The fast neutron irradiations were carried out behind the  $15 \times 15 \text{ cm}^2$  collimator of the cancer radiotherapy beam at the Institute of Nuclear Physics in Krakow. Here, fast neutrons are produced via the  $\text{Be}(d,n)\text{B}$  reaction from deuterons of 12.5 MeV accelerated by the U-120 cyclotron bombarding a thick Be target. The mean fast neutron energy is c. 5.6 MeV and the gamma contribution to the total (neutron + gamma-ray) dose is 4% (13). Fast neutron dosimetry for this beam is carried out according to the European Protocol (14). The total dose was measured by an Exradin chamber of  $0.53 \text{ cm}^3$  volume, in terms of Gy(tissue) per charge (Coulomb) of the deuteron beam, and the exposures controlled by measuring the beam charge (average beam current c.  $14.5 \mu\text{A}$ , calibration factor  $0.0112 \text{ cGy}/\mu\text{C}$ ). For neutron irradiations, ampoules containing the Fricke solution were exposed in air at the centre of the irradiation field. In two separate runs eight exposures were made in the range from 50 Gy to 200 Gy.

## 3. The model calculation

Track structure theory (11,12) relates the response of a detector after doses of "test" radiation ( $^{60}\text{Co}$  gamma rays) to its response after doses of heavy charged particles, specified by their charge, velocity and fluence (number of particles/ $\text{cm}^2$ ). In terms of this theory, the Fricke dosimeter is a 1-hit detector (10), described by three parameters: "c-hittdness" ( $c = 1$ ), characteristic radiosensitivity to test radiation,  $D_0$  ( $D_0 = 8000 \text{ Gy}$ ), and size of target given by its radius,  $a_0$  ( $a_0 = 6.5 \text{ nm}$ ).

The theory assumes that the signal  $S$  (e.g., the value of absorbance) of the Fricke dosimeter is linear with gamma-ray dose  $D_\gamma$  up to saturation, and given by the 1-hit formula:

$$S(D_\gamma) = S_S (1 - \exp(-D_\gamma/D_0)) \quad (3)$$

where  $S_S$  is the signal at saturation. The probability that a signal occurs after a dose of test radiation  $D_\gamma$ , is  $P(D_\gamma) = S(D_\gamma)/S_S$ .

It is assumed that the target is a sphere of water of radius  $a_0$  surrounding an  $\text{Fe}^{2+}$  ion such that a single hit (e.g., a transversal of one of the electrons, or delta-rays, surrounding the ion in its passage through the detector medium) can initiate the sequence of events which diffuse to the  $\text{Fe}^{2+}$  ion and finally transform it to an  $\text{Fe}^{3+}$  ion. The radial probability of this event occurring,  $P(r)$ , is calculated from the average energy deposited in the target volume (local dose),  $E_0(r)$ , calculated (9) as a function of the radial distance from the ion's path,  $r$ :

$$P(r) = P(E_0(r)) = P(1 - \exp(-E_0(r)/D_0)) \quad (4)$$

We calculate the single-particle action cross-section for this event  $\sigma$  by volume integration of the probability  $P(r)$  over all radii:

$$\sigma = 2\pi \int_0^T t P(t) dt \quad (5)$$

where the upper range of integration is limited to the maximum range of delta rays surrounding the ion,  $T$ .

After a fluence  $F$  particles/cm<sup>2</sup>, or ion dose, in water,  $D_i = F \times L$  ( $L$  is the stopping power of the ion, or its LET), the signal observed in a thin specimen (track-segment irradiation) is:

$$S(D_i) = S_S (1 - \exp(-\sigma F)) = S_S (1 - \exp(-\sigma D_i/L)) \quad (6)$$

The relative detector effectiveness (RE) is defined as the ratio of detector signals after equal doses of ion and test (gamma) radiations, i.e.:

$$RE = S(D_i)/S(D_\gamma) \quad ; \quad D_i = D_\gamma \quad (7)$$

After expanding equations (4) and (7) in the linear region and performing simple arithmetics, we arrive at the following

<sup>1</sup> Equation (2) in ref. (10) is erroneous

description of relative effectiveness in track segment irradiation:

$$RE \text{ (calculated)} = \sigma D_0 / L \quad (8)$$

This value can be compared with the measured G-values for ion irradiation of thin samples,  $G_i$ , and that for gamma-rays,  $G$  (for  $^{60}\text{Co}$ ,  $G(\text{Fe}^{2+}) = 15.6$ ):

$$RE \text{ (measured)} = G_i(\text{Fe}^{2+}) / G(\text{Fe}^{2+}) \quad (9)$$

To calculate the response of thick detectors to stopping ions, of initial kinetic energy  $T_i$  and of range  $R$ , we integrate the response over the path length. We have therefore to replace  $\sigma$  and  $L$  in eq. (9) by their average values:

$$\sigma_{\text{AVE}} R = \int_{t=0}^{t=R} \sigma \, dr = \int_{T_i}^0 (\sigma/L) \, dT \quad (10)$$

and:

$$L_{\text{AVE}} = T_i / R \quad (11)$$

The effect of irradiating the detector with a beam of fast neutrons is calculated by adding the contributions from all the charged secondaries stopping in the medium (11). Let us denote the number of neutron interactions per cubic centimeter of detector volume by  $Y$ , and the absorbed dose from neutrons by  $D_n$ .  $R_{z_i}$  represents the range (in cm) of an ion of atomic number  $Z$  and initial kinetic energy  $T_i$ , while  $dN_{z_i}/dT_i$  represents the number of secondary charged particles of atomic number  $Z$  and initial kinetic energy  $T_i$  per unit initial kinetic energy interval per neutron interaction per cubic centimeter of detector (i.e., the given secondary particle energy spectrum). Then:

$$D_n = Y \left[ \sum_Z \sum_{T_i} (\Delta T_i) (dN_{z_i}/dT_i) T_i \right] \quad (12)$$

and:

$$P(D_n) = 1 - \exp(-Y \left[ \sum_Z \sum_{T_i} (\Delta T_i) (dN_{z_i}/dT_i) \sigma_{\text{AVE},z_i} R_{z_i} \right]) \quad (13)$$

The detector signal after a neutron dose  $D_n$  is then  $S(D_n) = S_0 P(D_n)$ , and the relative effectiveness of the detector after a

neutron dose can be calculated from equation (8) and compared with experiment (eq. (10)) by replacing  $D_0$  by  $D_n$  and  $G_0$  by  $G_n$  (the measured G-value after a neutron irradiation) in these two equations.

The secondary spectra ( $dN_2/dT_0$ ) for all secondary particles generated in water after irradiation with the fast neutron beam of the MRC cyclotron in Hammersmith (private communication of Dr. John A. Dennis to Dr. Robert Katz, see also (16)) and the program for calculating the response of the Fricke dosimeter to this beam, have been kindly provided to one of the authors (MPRW) by Dr. Katz. The program, developed originally at Dr. Katz's laboratory (Lincoln, NE, USA) by Mrs. Rose Anne Roth for the IBM mainframe, has now been modified, also at Lincoln, by Mr. Gary L. Sinclair and Mr. Givargis Danialy to run on an IBM-PC/XT - compatible computer. The program is written in FORTRAN-77.

#### 4. Results

The results of measurements are given in Table 1 where the nominal dose,  $D_E$  (Gy), the run number, measured absorbance, temperature at which absorbance was measured, the calculated dose,  $D_0$ , and the ratio  $D_0/D_E$  are listed for the gamma-ray and fast neutron exposures. Values of molar extinction coefficient appropriate to the indicated temperature of measurement and  $G = 15.6$  were used for calculating  $D_0$ .

##### 4.1. Gamma-ray exposures

The average value of  $D_0/D_E$  for all seven points is  $\langle D_0/D_E \rangle = 1.027$ , the standard deviation  $SD = 0.078$  and the coefficient of variation ( $100\%SD/\langle x \rangle$ ) is  $CV = 7.6\%$ . Eliminating the highest and lowest measured values (points No. 4 and No. 6) does not appreciably change the average values:  $\langle D_0/D_E \rangle = 1.028$ , but the standard deviation decreases to 0.022 and the coefficient of variation  $CV = 2.2\%$ . We note that the value of  $\langle D_0/D_E \rangle$  greater than 1 indicates "underexposure", i.e. that the planned exposure is lower than expected, in terms of absolute units of dose.

##### 4.2. Fast neutron exposures

The average value of  $D_0/D_E$  is in this case  $\langle D_0/D_E \rangle = 0.582$ ,  $SD = 0.025$ ,  $CV = 4.38\%$ , if all eight points are considered. If one eliminates the highest and the lowest values of  $D_0/D_E$  in this set (points No. 4 and No. 5), the respective values are:  $\langle D_0/D_E \rangle = 0.585$ ,  $SD = 0.017$ , and  $CV = 2.96\%$ .

To calculate the G-value for the neutron component of our beam, where the contribution of neutron dose  $D_n$  to the total beam dose  $D_T$  is 96% as measured by tissue-equivalent ionization chambers (10), i.e.  $D_T = D_n + D_\gamma = 0.96 \times K \times D_T + 0.04 \times D_T$ , where  $K$  is the neutron kerma for water and  $D_\gamma$  is the gamma-ray dose contribution to the total beam dose  $D_T$ , we calculate the number of  $Fe^{2+}$  ions produced by each component of the beam:

For gamma-rays:  $N_\gamma = G \times D_\gamma$  (we assume  $G = 15.6$ )  
 For neutrons:  $N_n = G_n \times D_n$

The total number of  $Fe^{2+}$  ions is then:

$$N_T = G_n \times K \times 0.96 \times D_T + G \times 0.04 \times D_T \quad (14)$$

By rearrangement:

$$G_n = [(N_T/D_T) - G \times 0.04] / (0.96 \times K) \quad (15)$$

We note that  $N_T/D_T = G \times D_C/D_E$ , where  $D_C/D_E$  (for neutron exposures) is the value measured experimentally. Finally,

$$G_n = G \times [(D_C/D_E) - 0.04] / (0.96 \times K) \quad (16)$$

We calculate the value of  $K$  as the ratio of kerma for water to kerma for tissue at 5.7 MeV neutron energy from the tables of Bach and Caswell (17):  $K = 1.094$ . The G-value for our fast neutron beam is then:

$$G_n = 15.6 \times (0.585 - 0.04) / (0.96 \times 1.094) = 15.6 \times 0.52 = 8.10$$

The error on this value can be estimated as a sum of the relative errors of evaluating  $D_C/D_E$  (3%), and of experimental dosimetry for gamma-rays (2%) and for the neutron beam (2%). Thus:

$$G_n = 8.1 \pm 0.6$$

#### 4.3. Model calculations

The program has been run on an 8 MHz IBM-PC/XT compatible computer equipped with an 8087 mathematical co-processor and 640 Kb RAM memory. When compiled using the Ryan-McFarland Profort V.1.0 compiler, the program completes the calculation in about 3 hours. The secondary particle spectra are for the Hammersmith fast neutron beam irradiating pure water. The Fricke detector was represented by the following parameters (units are those used in the program):  $C = 1$ ,  $E_0 = 8 \times 10^8$  erg/cm<sup>3</sup>,  $a_0 = 4.5 \times 10^{-7}$  cm. The calculated efficiency of this detector, relative to gamma-rays, is  $\epsilon = 0.552$ . The calculated G-value is therefore:

$$G_n = 15.6 \times 0.552 = 8.61$$

## 5. Discussion

The results of this work raise several questions. We judge the overall reproducibility of our measurements (our random uncertainty is at best 3%) to be inferior to that quoted in the literature, of 1% (1,2).

We believe that this poor reproducibility is mainly due to the insufficient purity of the sulphuric acid used in our dosimeters. It appears that special purification is required for our application. Particular attention has also to be paid to minor inorganic contaminations on the inner walls of the ampoules, which can possibly alkalize the dosimetric compound.

The calibration of the spectrophotometer appears to be quite satisfactory, though care has to be taken to read absorbances only within the range of the calibration curve. We have eliminated from our analysis gamma-ray exposures below 40 Gy for that reason.

Despite the large uncertainty in the results of gamma-ray exposures, there is an indication of a systematic "underexposure", of about 3%. Interestingly, a similar result was obtained when exposing alanine detectors to the same beam (15). These results may indicate the presence of a systematic difference of about -3% in the absolute gamma-ray dosimetry at the Centre of Oncology in Krakow, or in relation to, e.g., Risoe National Laboratory, Denmark. While the discrepancy is well within experimental error, its observation in two independent experiments using two different dosimeters, could perhaps be significant.

The results of Fricke dosimetry performed at the MRC Hammersmith beam, quoted by Green et al. (8), when presented in terms of the neutron  $G_n$ -value, are  $9.74 \pm 0.10$  (17 irradiations) and  $9.59 \pm 0.07$  (18 irradiations) where the errors are random only. Taking into account their overall uncertainty of 6% due to basic ionization dosimetry, Green et al. arrive at a mean value of  $G_n = 9.4 \pm 0.6$ .

We have yet to meet this repeatability, however, the difference between our result ( $G_n = 8.1 \pm 0.6$ ) and that obtained at Hammersmith clearly indicates that the differences between the neutron spectra at Hammersmith and in Krakow are significant enough to be resolved by Fricke dosimetry. We have confirmed this result in our alanine work (15), where our measured relative effectiveness was  $0.60 \pm 0.05$ , while that reported by Simons and Bewley (19) for the Hammersmith beam is  $0.65 \pm 0.05$ . The gradual increase in the  $G_n$ -value with increasing neutron energy (for mono-energetic neutrons) has been studied by Lawson and Porter (5). Our result is quite compatible with their evaluations.

The value of  $G_n$  calculated from track structure theory, though, as

expected, larger than our measured value, underestimates the Hammersmith result (the calculated value is 6% above ours and 12% below the MRC value). Interestingly, while track structure calculations with the same values of the three parameters appear to be consistent with the reported differentiation yields in the Fricke dosimeter for charged particles (corresponding to track segment irradiation) to within about 5 %, the same calculation underestimates the reported integral yields (after stopping particles), also by about 15 % (10). This could indicate problems with the stopping particle model at low energies, a major part of the neutron calculation.

One must attribute these discrepancies mainly to uncertainties in our knowledge of the "effective charge", of the radial distribution of dose and of the stopping power and range at low energies of heavy ions.

Some calculational error could also be introduced through errors in the determination of the energy spectra of charged secondaries from the fast neutron beam in water and through the assumption in our calculations that the Fricke dosimeter is composed of pure water only.

The need to evaluate secondary particle spectra for the U-120 neutrons seems to be evident. First, however, the neutron energy spectrum of the Krakow beam would have to be evaluated, a problem of some complexity.

For radiobiological purposes, however, the presently obtainable calculational accuracy of about 10% would perhaps not be unreasonable.

## 6. Conclusions

Ferrous sulphate dosimetry is an excellent and highly accurate technique if the high demands for compound purity and chemical standards are met. Our first attempt, though in need of further improvement, has demonstrated the usefulness of this method. Together with an independent tissue-equivalent dosimetric system - alanine - and a theoretical model which presently reproduces the experimental results for the Fricke and alanine dosimeters to within about 10% (but perhaps could be improved), we are able to build a consistent dosimetric system for measuring fast neutron doses. The present reproducibility of our Fricke dosimetry is no worse than 4%. Though further work is needed to improve the accuracy of our Fricke dosimetry and of our calculations, the system can already be used for fast neutron radiobiology at the U-120 cyclotron. The observed indication of a possible discrepancy in the absolute dosimetry of gamma-ray doses at the Centre of Oncology in Krakow will need further confirmation and corrective work if it is found to be real.



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Table 1. Experimental results of Fricke dosimetry in the gamma-ray and the fast neutron beams

<sup>60</sup>Co gamma-rays

Point No.	Run No.	Exp. dose <sup>1</sup> D <sub>E</sub> , Gy	Absor- bance cm <sup>-1</sup>	temp. of meas. deg C	Calc. dose <sup>1</sup> D <sub>C</sub> , Gy	D <sub>C</sub> /D <sub>E</sub>
1	1	98.29	0.340	18.5	97.91	0.996
2	1	147.18	0.536	18.5	154.35	1.049
3	1	196.07	0.715	18.5	205.90	1.050
4	2	49.66	0.211	24.5	58.32	1.174
5	2	49.66	0.172	24.5	47.54	0.957
6	2	99.33	0.337	24.5	93.15	0.938
7	2	99.33	0.369	24.5	102.00	1.027

U-120 fast neutrons

Point No.	Run No.	Exp. dose <sup>2</sup> D <sub>E</sub> , Gy	Absor- bance cm <sup>-1</sup>	temp. of meas. deg C	Calc. dose <sup>3</sup> D <sub>C</sub> , Gy	D <sub>C</sub> /D <sub>E</sub>
1	1	89.38	0.187	18.5	53.85	0.602
2	1	89.38	0.189	18.5	54.43	0.609
3	2	50.00	0.103	24.5	28.47	0.569
4	2	50.00	0.097	24.5	26.81	0.536
5	2	150.00	0.332	24.5	91.77	0.612
6	2	150.00	0.317	24.5	87.62	0.584
7	2	200.00	0.412	24.5	113.88	0.569
8	2	200.00	0.415	24.5	114.71	0.574

<sup>1</sup> absorbed dose, in water

<sup>2</sup> absorbed total beam dose, in tissue

<sup>3</sup> assuming G = 15.6 and D<sub>E</sub> = D<sub>γ</sub> (100% gamma, in water)