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Dosimetry of Fast Neutrons from T (d, n) Reaction Neutron Generator

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T (d,n) 反応中性子線の線量測定

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Cockcroft-Walton 型 T(d, n) 中性子線の線量を Fricke 線量計, $^{32}\text{S}(n, p)^{32}\text{P}$ 反応による放射化法ならびにプラスチック・シンチレータによる蛍光法によつて測定した。

Fricke 線量計とイオウの放射化法による値は良い一致を示したが, シンチレーション・カウンタと他の2つの線量計の間には, そのような相関はみられなかつた。これはターゲット周辺の中性子束分布が, 重水素イオンビーム・スポットのターゲット上における位置, 大きさや加速電圧な

らびにターゲットの使用歴によつて変るので, ターゲットに対する線量計の幾何学的条件の違いに基づくものであることがわかつた。

この結果, シンチレーション・カウンタは大凡の線量制御には有用であるが, 正確な線量は照射試料と幾何学的に同じかまたは似た状態の線量計によつて決定されなければならないことが結論された。現状では, イオウの放射化法が最も良いと思われる。

Introduction

Treatment of malignant tumors by fast neutrons has recently been initiated at some institutions in Japan. The prospects of greater use of neutrons in the near future have brought about a need to develop practical dosimeters for measuring fast neutrons. Standard methods for measuring X- and γ -rays, and electrons have been established and reported by Japanese Association of Radiological Physicists, Japan Radiological Society¹⁾²⁾, but such a standard method for measuring neutrons has not yet been developed, probably because of the paucity of neutron sources available in Japan. In order to establish procedures suitable for neutron dosimetry in clinical radiology and radiobiology, intercomparison of neutron dosimetry among institutions equipped with Cockcroft-Walton type T(d,n) neutron generator was made using the

activation method of $^{32}\text{S}(\text{n,p})^{32}\text{P}$ reaction. Unlike Co-60 γ -rays, high energy X-rays and electrons, results from repeated measurements of neutrons varied widely among institutions and even within institution.

In the present study, values obtained by the activation method and scintillation method routinely used in our institute were compared with those obtained by a Fricke dosimeter, and the factors affecting dose estimations were reviewed.

Materials and Methods

Fast neutrons were produced by bombarding a tritium target with deuterons accelerated at 100–180 kV. Deuteron beam current was 1.2 mA.

Dosimeters used were a single channel scintillation spectrometer equipped with a plastic phosphor (1 inch $\phi \times$ 1 inch, styrene phosphor), sulfur (guaranteed reagent, in 1.3 cm $\phi \times$ 3 cm polystyrene container) and a Fricke dosimeter (10^{-3}M $\text{Fe}(\text{NH}_4)_2(\text{SO}_4)_2$, 10^{-3}M NaCl , 0.8N H_2SO_4 in 1.3 cm $\phi \times$ 3 cm pyrex glass container). The irradiated sulfur samples were burnt on a sand bath and the activity of the residual ^{32}P was measured by a GM counter whose counting efficiency had been determined previously by a standard ^{32}P source. A value of 248 mb^{83} was employed as the nuclear reaction cross section of $^{32}\text{S}(\text{n,p})^{32}\text{P}$ reaction for 14.1 MeV neutrons, and for changes in cross section by neutron energy, corrections were made when necessary⁴⁾⁵⁾. Neutron flux from radioactivity of ^{32}P and counts of recoil protons produced in the plastic phosphor was calculated by methods reported previously⁶⁾⁷⁾. A factor of $6.94 \times 10^{-9} \text{ rad/n}\cdot\text{cm}^{2\ 83}$ was used in converting neutron flux to kerma for 14.1 MeV neutrons, assuming that Fricke dosimeter is equivalent to water. Errors by neglecting the variation of the factor by neutron energy were estimated to be less than 5%. The G-values of the Fricke dosimeter for 14 MeV neutrons are reported to range from a minimum of 8.3 to a maximum of $11.8^{9)-14)}$. Law et al.¹¹⁾ have recommended a value of 9.9 ± 0.6 for T(d,n) neutrons and ^{252}Cf neutrons including contaminated γ -rays. The γ -ray dose in the neutron fields measured by CaSO_4 thermoluminescent dosimeter was 3.7%, comparable to those reported by Law et al.¹¹⁾ Therefore, a value of 9.9 was used in the present study. Ferric ions were quantitated using quartz cells 5 cm in length to detect low neutron doses, because of lower G-value for neutrons than γ -rays and the high cost of tritium target.

For relative measurements, $^{56}\text{Fe}(\text{n,p})^{56}\text{Mn}$ reaction was used. The resonance peak of the reaction appears at 14 MeV and thus variations of the reaction cross section by neutron energy are small, when compared to $^{32}\text{S}(\text{n,p})^{32}\text{P}$ reaction.

Plastic phosphor was placed at 30° right of and 100 cm from the target. Sulfur and Fricke dosimeters were located 5 cm around the target, the distance routinely used for irradiation of biological materials.

Results

1. Intercomparison of T(d,n) neutrons by sulfur activation detector

Seven sulfur samples were mailed to Kyushu and Nagasaki Universities equipped with T(d,n) reaction neutron generator, and six samples were irradiated with neutrons of about 100 rads under designated technical factors. The samples when returned were treated similarly to those irradiated at our institute, and their radioactivity was measured. The results from three repeated comparisons are summarized

Table 1 Intercomparison of T (d, n) reaction neutrons by sulfur activation detector

Experiment No.	Evaluated/Exposed (mean, σ)		
	Institution I	Institution II	Institution III
1	0.75, 0.02	1.11, 0.05	1.15, 0.03
2	0.79, 0.08	0.97, 0.04	0.98, 0.06
3	0.84, 0.08	1.07, 0.05	1.01, 0.07

Monitors used were Si (Li) semiconducting α -detector in institutions I and II, and scintillation counter in institution III.

in Table 1. Neutron monitors used were Si(Li) semiconducting α -detector in institutions I and II, and scintillation counter in institution III, our institute. As shown in the table, wide variations were observed among institutions and within institution. In relative measurement, the accuracy of neutron flux measurement by sulfur activation detector was within 5%, and the deviations observed exceeded experimental errors.

2. Fricke dosimetry

By the use of quartz cells 5 cm in length, it was possible to make the determination of γ -ray doses exceeding 800 rads within a standard deviation of less than 2%. As the path of ultra-violet rays across the cells was longer than usual, the accurate setting of the cells in measuring UV absorption was important. In view of low G-value for neutrons, Fricke dosimeter was exposed to about 1500 rads.

The values obtained by Fricke dosimeter and sulfur detector in comparison with those by scintillation counter used as a monitor are shown in Figure 1. In seven repeated experiments, the values of sulfur detector agreed well with those of Fricke dosimeter. However, such an agreement could not be observed between the values of scintillation counter and the two other dosimeters.

Changes in sensitivity of the scintillation counter to neutrons and differences in geometric factors of

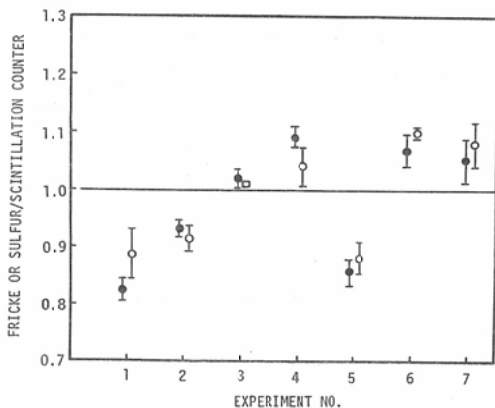


Fig. 1. Comparison of neutron doses measured by Fricke dosimeter, sulfur activation detector and scintillation counter

● Fricke dosimeter, ○ Sulfur activation detector

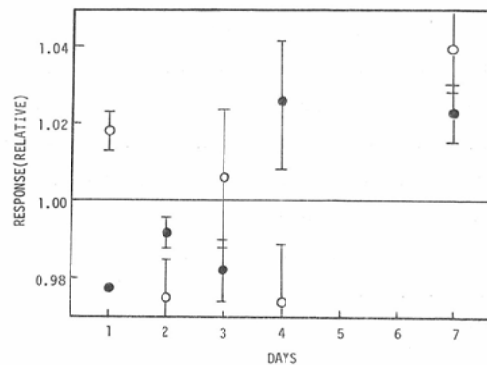


Fig. 2. Daily change in response of scintillation counter

Source: $^{241}\text{Am-Be}$ neutron source

● Summer, ○ Winter

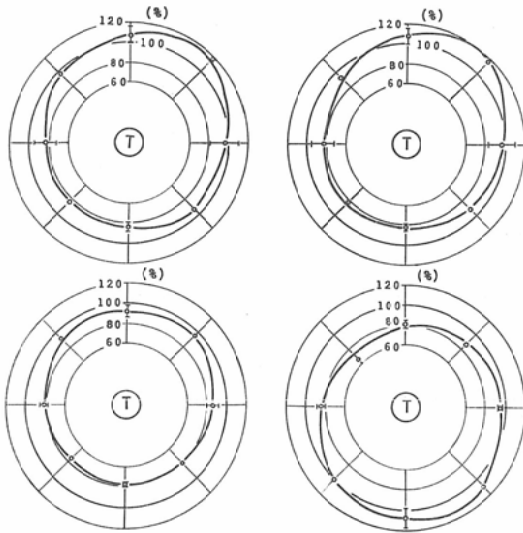


Fig. 3. Angular distribution of neutron flux density on plane perpendicular to deuteron beam

the dosimeters may be the responsible causes for the disagreement among the three dosimeters.

3. Daily changes in response of the scintillation counter

The daily changes in response of the scintillation counter to neutrons from a $^{241}\text{Am-Be}$ source were studied for one week. As shown in Figure 2, daily changes were within 6%. This could not explain the differences in dose measured by the counter and Fricke dosimeter.

4. Distribution of neutron flux density around the tritium target

Figure 3 shows the neutron flux density distributions on the plane perpendicular to deuteron beam, measured by activation of $^{56}\text{Fe}(n,p)^{56}\text{Mn}$ reaction. The results of four experiments at different days are shown separately. Significant daily changes in focal spot of deuteron beam on the tritium target were observed.

Those on the plane parallel to deuteron beam by accelerating voltage are shown in Figure 4. Neutron flux density at the right of the target showed a tendency to increase at higher voltage, indicating that location and size of the beam spot differ by voltage. Tritium target age was another factor influencing these distributions in addition to accelerating voltage. The neutron output from the outer area of the target in comparison to that from the central area increased by consumption of tritium at the central area of the target due to continuous bombardment of deuteron beam.

As mentioned above, neutron flux density around the target was dependent of the location of beam spot, accelerating voltage and target age, but these were difficult to maintain constant in routine exposures. These were found to be the factors responsible for variation of neutron doses measured by scintillation counter at 100 cm from the target and Fricke dosimeter near the target. This may be the case for $\text{Si}(\text{Li})$ α -detector used in other institutions.

Discussion

$\text{T}(d, n)$ neutron generators are being used widely in Japan because they are compact, easy to operate

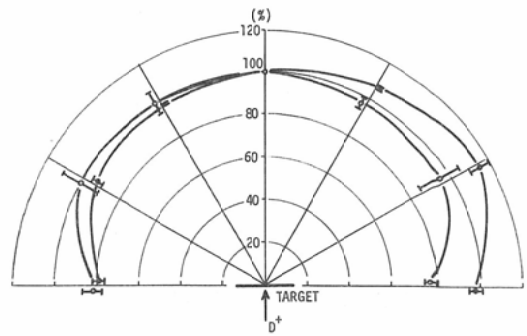


Fig. 4. Angular distribution of neutron flux density on plane parallel to deuteron beam
○ 100 kV, ● 160 kV

and can generate monochromatic neutrons. However, output from the generators are relatively low for radiobiological studies and clinical radiology, and thus irradiating materials must be placed close to the target for exposures of several hundred rads.

Scintillation counter based on proton recoil measurements is advantageous in that the dose rate and dose during irradiation can be determined, but it is too sensitive as a neutron dose monitor. The distance of less than 100 cm between phosphor and target brought about errors due to counting loss and pile up of recoil protons produced in phosphor. The value obtained by the scintillation counter was 1.85×10^{-5} rad/count, and consistent with that by Fricke dosimeter¹⁵⁾ when the distribution of neutron flux density was nearly isotropic. Even though the counter was operating normally, the differences in neutron flux density near the target did not reflect proportionally to the scintillation counter. The scintillation counter in our institute was compared to tissue equivalent chambers at the National Institute of Radiological Sciences which participated in and was calibrated by international comparison. However, the results were not satisfactory for reasons mentioned above.

For T(d,n) neutron generator, doses to samples should be measured by dosimeters located at the same or similar geometric conditions to the samples unless the generator produces stable neutron flux. At present stage, sulfur detector is the most convenient one for T(d,n) neutrons, because the size and shape of the detector can be easily changed.

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Summary

Neutron doses from a Cockcroft-Walton type T(d, n) neutron generator were measured using a Fricke dosimeter, activation of $^{32}\text{S}(n, p)^{32}\text{P}$ reaction and a scintillation counter with a plastic phosphor.

A good agreement in doses measured by Fricke dosimeter and sulfur detector was observed but such an agreement was not noted between scintillation counter and the two other dosimeters. This was due to differences in geometric conditions of dosimeters relative to the tritium target because neutron flux density around the target varied by the location and size of deuteron beam spot, accelerating voltage of deuterons and target age.

It was concluded that scintillation counter is useful for tentative dose control, but accurate doses to samples should be determined by dosimeters placed in the same or similar geometric conditions to those of the samples.

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