Dose Measurement of $^{90}$Sr Source for Radiation Chemical Reaction

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The irradiation characteristics for a plate source of $^{90}$Sr 222 GBq (6 Ci) were examined with the blue cellophane dosimeter and the thermoluminescence dosimeter. The $^{90}$Sr source used has the active area of $35 \times 20 \text{mm}^2$ and the surface concentration of $31.5 \text{GBq (0.85 Ci)/cm}^2$. The high dose rate of $1.1 \text{kGy (1.1 X 10^5 rad)/h}$ was obtained at the distance of 5 cm from this source. At that time, the isodose curve determined by the blue cellophane was similar to a circle. The peak in depth dose curve was fairly displaced to the shallow direction in comparison with that obtained from the irradiation with an electron beam accelerator (2 MeV). The calculated dose rate attains to $800 \text{kGy (8.0} \times 10^7 \text{rad)/h}$ at the distance of 5 cm from the source when the infinite plate source with the maximum surface concentration ($1.51 \text{TBq (40.7 Ci)/cm}^2$) is placed on a plane.

Key Words: dose measurement, plate source, strontium-90, isodose curve, depth dose curve

1. Introduction

Since $^{90}$Sr is a $\beta$-emitter which has a relatively long half-life (28 y) and high energy $\beta$-rays ($E_{\text{max}}=0.544, 2.27 \text{MeV}$), it has a bright prospect of the utilization as a radiation source for radiation process by making use of the characteristics of source. The utilization method can be classified into two systems: the internal source system for the chemical reaction in the liquid phase and the external source system for the surface treatment in the solid phase. In several foreign countries, the development of the irradiation facility of two systems has been made actively. Examples of the internal source system are the addition reaction facility using a tubular source (telomerization of methanol with ethylene) and the chlorination reaction facility using many microsphere sources (toluene chlorination). On the other hand, an example of the external source system is the graft reaction facility using a plate source (grafting on polypropylene). However, Japan is slow in the development of these two systems.

Thus, as a part of the development for the wide utilization of fission products, we studied the characteristics of $^{90}$Sr as a radiation source for the radiation chemical reaction (the surface treatment of solid body). Three plate sources of 222 GBq (6 Ci) were obtained from the Radiochemical Centre Ltd. and the irradiation facility was designed and built.

In order to investigate the irradiation characteristics of one of the three sources, the measurement of dose was carried out with the blue cellophane (BC) dosimeter and the thermoluminescence (TL) dosimeter. The relationship between dose rate and distance from the source, the spatial dose distribution, the depth dose distribution, and the effect of base materials of irradiated object on dose rate were examined. In addition, the dose rate from an infinite plate source was estimated and the feasibility of a radiation source for the surface treatment of solid body was discussed.

2. Experimental

2.1 $^{90}$Sr source

The structure of $^{90}$Sr source is shown in Fig.
1. The overall dimensions are $55 \times 40 \times 5 \text{ mm}^3$ (active dimensions: $35 \times 20 \text{ mm}^2$). The surface concentration of radioactivity for this source is $31.5 \text{ GBq (0.85 Ci)}/\text{cm}^2$. Strontium titanate SrTiO$_3$ incorporated in silver foil (with thickness of 0.05 mm) is further encapsulated in stainless steel case with a window of 0.05 mm in thickness. A backing piece for the silver foil is made from graphite to reduce the bremsstrahlung caused by $\beta$-rays.

2.2 Making of calibration curve for dosimeter

The blue cellophane ($45 \times 13 \times 0.025 \text{ mm}^3$) made in Avisco Ltd. and the LiF-teflon (UT-LiF-7, $6\times 0.02 \text{ mm}^3$) made in Teledyne Isotopes Ltd. were used for the dosimeters for $\beta$-rays. For the calculation of the absorbed dose, the calibration curves for both dosimeters (BC dosimeter: $10 \sim 150 \text{ kGy (1$\times 10^6 \sim 1.5 \times 10^7 \text{ rad}$)), TL dosimeter: $30 \sim 700 \text{ Gy (3$\times 10^3 \sim 7 \times 10^4 \text{ rad}$)}) were previously made up based on exposure to $^{60}$Co $\gamma$-rays. In the $\gamma$-ray irradiation of the dosimeter, the secondary electrons produced by $\gamma$-rays were equilibrated in the shielding material; acrylic resin plate with the thickness of 2 mm was used in the case of BC dosimeter, and teflon plate with the thickness of 4 mm was used in the case of TL dosimeter. The absorbed dose $D_{BC}$ of blue cellophane and the absorbed dose $D_{TL}$ of LiF-teflon are expressed as follows, respectively.

$$D_{BC} = D_A \cdot \frac{m_{SW}}{m_{SW}} = 0.869X \cdot \frac{m_{\beta A}}{m_{\beta A}} \cdot \frac{m_{SW}}{m_{SW}}$$

$$D_{TL} = D_T \cdot \frac{m_{STL}}{m_{STL}} = 0.869X \cdot \frac{m_{\beta T}}{m_{\beta T}} \cdot \frac{m_{STL}}{m_{STL}}$$

where $D_A$, $D_T$ are the absorbed dose of acrylic resin plate and the absorbed dose of teflon plate, respectively; $\frac{m_{SW}}{m_{SW}} = 0.972$, $\frac{m_{STL}}{m_{STL}} = 0.994$ the ratio of the average mass stopping power of blue cellophane to acrylic resin plate and the ratio of the average mass stopping power of LiF-teflon to teflon plate, respectively; $X$ the $\gamma$-ray exposure, and $m_{\beta A}$, $m_{\beta T}$, $m_{\beta A}$, $m_{\beta T}$, the mass energy absorption coefficient for $^{60}$Co $\gamma$-rays in acrylic resin plate, teflon plate and air, respectively.

The degree of fading of the BC dosimeter after irradiation was determined by measuring change in the optical density at 665 $m_{\mu}$ with a Narumi Ltd. microphotometer. The quantity of the thermoluminescence of the TL dosimeter was measured with a Teledyne Isotopes TLD-7300 B reader.

2.3 Measurement for absorbed dose

In the dose measurement of $^{90}$Sr source, the dosimeter was suspended by a thread at the fixed position and the source was set on a supporter. In the case of the BC dosimeter, the absorbed dose was calculated according to the following procedure. The absorbed dose $D_{BC}$ of blue cellophane was obtained from the decrease in optical density which takes place with the irradiation of $^{90}$Sr $\beta$-rays, using the calibration curve made up by $^{60}$Co $\gamma$-rays (in Sec. 2.2). Since the dependence of the response on the type and the quality of radiation are not recognized at the energy of 0.3$\sim$3 MeV, the calibration curve made up by $^{60}$Co $\gamma$-rays was applied for both dosimeters$^{5} \sim 8$). Then, $D_{BC}$ was converted into the absorbed dose $D_w$ of water using equation (3). In the case of the TL dosimeter, equation (4) was used for converting $D_{TL}$ into $D_w$.

$$D_w = D_{BC} \cdot \frac{m_{SW}}{m_{SW}}$$

$$D_w = D_{TL} \cdot \frac{m_{SW}}{m_{STL}}$$

where $\frac{m_{SW}}{m_{SW}} = 1.075$ is the ratio of the average mass stopping power of water to blue cellophane, and $\frac{m_{SW}}{m_{STL}} = 1.237$ the ratio of the average mass stopping power of water to LiF-teflon.
3. Experimental Results and Discussions

3.1 Relationship between dose rate and distance from source

The dose rate distribution along the axis ($z$) of source is shown in Fig. 2. Since the measurement must be carried out over a wide range of dose rate $10^{-10}$ to $10^{0}$ Gy ($10^{3}$ to $10^{6}$ rad)/h, the BC dosimeter was set at five points at the distance of $0$ to $5$ cm from the source and the TL dosimeter was set at seven points at the distance of $2.5$ to $50$ cm from the source. The calculated values in the figure were obtained by integrating numerically Loevinger’s empirical formula (5) for the beta point source, and by correcting for backscatter (graphite) $f_{1}$, self-absorption $f_{2}$ and absorption due to coating materials (Ag, stainless steel) $f_{3}$:

$$I(z) = \frac{1.7 \times 10^{6} \rho \nu E_{av}}{z^{2} [3c - (c^{2} - 1)] e} \left[ c \left( 1 - \frac{\nu z}{c} \exp \left( 1 - \frac{\nu z}{c} \right) \right) + \nu z \exp (1 - \nu z) \right]$$

where $I(z)$ is the dose rate at the distance of $z$ cm in air from the source (rad/h), $\rho$ the mass density of air (=0.001293 g/cm$^3$), and $E_{av}$ the average beta energy (MeV). The variable $\nu$ is called the apparent absorption coefficient (cm$^2$/g) and is given by the formula (6) (in air):

$$\nu = \frac{16.0 \times 10^{6}}{(E_{max} - 0.036)_{1+\nu}} \left[ 2 - \left( \frac{E_{av}}{E_{av}^{*}} \right) \right]$$

where $E_{av}^{*}$ is the hypothetical average beta energy per disintegration for a hypothetical allowed spectrum having the maximum energy value $E_{max}$ (MeV). The parameter $c$ is a dimensionless parameter and can be obtained from formula (7) (in air):

$$c = 3.11 \times \exp (-0.55 E_{max})$$

As can be seen from Fig. 2, the dose rate decreases rapidly with increasing the distance from the source. The experimental values agree relatively well with the calculated values in the vicinity of the source, while the former become higher than the latter in the distance away from the source. The reason for this is considered as the energy dependence of the TL dosimeter (10). That is to say, since the $\beta$-rays scattered in air layer between the source and the dosimeter increase with distance from the source, the energy is wholly displaced to the low energy region, and consequently the readings of the TL dosimeter are higher than the true values, because the response is higher in the low energy region (below 100 eV). However, the detail is not obvious. The dose rate of 1.1 kGy ($1.1 \times 10^{3}$ rad)/h was obtained at the distance of 5 cm from the source.

3.2 Spatial dose distribution

The spatial dose distributions at the distance of 1 and 5 cm from the source are shown in Figs. 3 and 4. The BC dosimeter having a size of $120 \times 160$ mm$^2$ was used and the degree of fading was measured by scanning continuously with the microphotometer. The numerical values are expressed in the ratio to the maximum dose. In the vicinity of source ($z=1$ cm), the isodose curve was extremely distorted and the spacings in all the two isodose curves expanded as the distance from the source center increased. The curve was similar to the shape of source. On


3.3 Depth dose distribution

The depth dose distribution at the distance of 2.5 cm from the source is shown in Fig. 5. Irradiation was carried out with stacking alternately the BC dosimeter and teflon plate. The dose on the surface of incidence was 74% of the maximum value. The curve had a peak at the thickness of 0.073 g/cm² and the effective path $R_{ef}$ was 0.19 g/cm². The peak in this curve was fairly displaced to the shallow direction in comparison with depth dose curve obtained from irradiation with an electron beam accelerator (2 MeV). In explanation of this, it is considered that $^{90}$Sr $\beta$-rays were absorbed with the coating materials (Ag, stainless steel) of the source and consequently the shallow part of the depth dose distribution curve disappeared. In this source, since the incident electrons have the distribution in the direction and the energy, the influence of the absorption appeared highly compared with the case of an electron beam accelerator.

3.4 Effect of base material of irradiated object on dose rate

When the base material is placed behind the irradiated object, the dose rate is affected by the backscatter from the base material. Therefore, the effect of the base materials was examined. Wood, teflon, aluminum, iron, tin and lead were used for the base materials. The thickness of the base materials is thick enough to saturate the $\gamma$-ray backscatter factor. Irradiation was carried out with setting the BC dosimeter on the base material. The relationship between the backscatter factor and atomic number of the base material is shown in Fig. 6. The backscatter factor increased with atomic number $Z$ of the base material. The backscatter factor obtained by this experiment (dose measurement method) was greater compared with the value of Burtt obtained by the radiation
counting method (over Z=40). This cause is considered as the difference in the measurement for the backscatter factor, and further the energy dependence of the BC dosimeter for the β-rays scattered by the base material.

3-5 Estimation of dose rate from infinite plate source

The industrial utilization of the 90Sr source makes it necessary to investigate the dose rate from the plate source of wider active area and higher surface concentration. Therefore, the dose rate $I_a(z)$ from an infinite plate source was evaluated from the formula (8):

$$I_a(z) = \frac{1.07 \times 10^6 \mu E_\alpha \sigma}{3 \rho^2 - (\rho^2 - 1)x} \cdot \left[ e \left( 1 + \ln \frac{e}{\nu \Delta z} - e^{-\nu / \Delta z} \right) + e^{-\nu / \Delta z} \right] \cdot f_1 \cdot f_2 \cdot f_3$$

(8)

where $\sigma$ is the surface concentration (Ci/cm²).

In Fig. 7 is shown the result of the infinite plate source (source B) together with the result of the present 222 GBq (6 Ci) source (source A). In this case, the dose rates from the both sources with a surface concentration of 31.5 GBq (0.85 Ci)/cm² were compared. The dose rate from the infinite plate source decreases gradually. The dose rate of 62 kGy (6.2×10⁶ rad)/h can be obtained at the distance of 5 cm from the source B.

Next, the dose rate from the source with a surface concentration of 1.51 TBq (40.7 Ci)/cm² (theoretical maximum value) was estimated. This maximum surface concentration is determined from multiplying maximum specific activity (1.41 TBq (38.0 Ci)/g) by maximum range (1.07 g/cm²). Table 1 shows the comparison of the dose rates from the sources with the various characteristics. In this case, the active area (7 cm² and infinite) and the surface concentration (31.5 and 1510 GBq (0.85 and 40.7 Ci)/cm²) were adopted as the characteristics. The dose rate at the distance of 5 cm from source C (surface conc.: 1.51 TBq (40.7 Ci)/cm², active area: ∞) becomes 800 kGy (8.0×10⁷ rad)/h which is about 700 times as large as for the source A. This dose rate can not compete well with the electron beam accelerator. However, if the source is processed to the same shape as the irradiated object, it will be applicable to the surface treatment of bodies with the complicated shape which is difficult to irradiate with the electron beam accelerator.
4. Conclusion

The irradiation characteristics for a plate source of \(^{90}\text{Sr} \) 222 GBq (6 Ci) were examined with the BC dosimeter and the TL dosimeter. In the vicinity of the source, the dose rate became fairly high and the isodose curve was similar to the shape of the source. The dose rate decreased rapidly with distance from the source and the isodose curve was nearly a circle. The dose rate of as high as 800 kGy (8.0 \( \times 10^7 \) rad)/h will be obtained at the distance of 5 cm from the sources when the sources with a surface concentration of 1.51 TBq (40.7 Ci)/cm\(^2\) are placed on an infinite plane. Thus, if the \(^{90}\text{Sr} \) source is processed to the same shape as the irradiated object, it will be applicable to the surface treatment of bodies having the complicated shape.

References

1) P.M. Yavorsky, et al.: NYO-2978-42 (1965)
7) J. Harvey and S. Townsend: RD/B/N 829 (1967)
10) G.W.R. Endres: BNWL-339 (1965)
12) B.P. Burtt: *Nucleonics*, 7, August, 29 (1948)

要 旨

放射線化学反応用 \(^{90}\text{Sr} \) 線源の線量測定

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\(^{90}\text{Sr} \) 222 GBq (6 Ci) の板状線源の照射特性を調べるため、プレーゼロファン線量計と熱ルミネセンス線量計を用いて、その線量測定実験を試みた。使用した \(^{90}\text{Sr} \) 線源の放射面積は 35 \( \times \) 20 mm\(^2\) で、その放射能面密度は 31.5 GBq (0.85 Ci)/cm\(^2\) である。
線源から 5cm 離れた点では、1.1 kGy (1.1 \( \times 10^8 \) rad)/h の高い線量率が得られ、等線量線は円形に近かった。深部線量線は電子線加速器 (2 MeV) による照射で得られるそれに比して、ピークの位置が浅い方へかなり変位していた。最大放射能面密度 1.51 TBq (40.7 Ci)/cm\(^2\) に有する線源を無限板状に配置すると、線源から 5cm 離れた点の線量率は 800 kGy (8.0 \( \times 10^7 \) rad)/h に達する。