# Gamma-Ray Dosimetry in an Irradiation Cavity of the 2000 Curie Cobalt-60 Facility

Sakae SHIMIZU, Shin TANAKA, and Yasuyuki NAKAYAMA*

Shimizu Laboratory, Institute for Chemical Research, Kyoto University

For the 2000 Curie Co⁶⁰ irradiation facility installed in our Institute the measurment and calculation of dose rates in its irradiation cavity were carried out. The Fricke ferrous sulfate dosimeter, utilizing the ferrous-ferric reaction by the effect of gamma-rays, was applied to the experimental work, as its good characteristics seemed to be favorable for the present purpose. While a few Ag-activated phosphate glass needles were also used in parallel with the chemical dosimetry. The mathematical calculation of dose rates basing upon several assumptions was performed to compare those with the experimental results. The comparison showed that experimental results were in reasonably good agreement with calculated values. The discrepancy was presumably due to the fact that the dose rates given by the Fricke dosimeters were the spatially averaged values of gamma-ray intensities in the regions occupied by the dosimeter solutions, and due to several assumptions on which the mathematical calculation was performed. Considering many conditions, the dose rate at the center of the cavity was determined with a probable error of about 5 percent. It can be mentioned that the Fricke chemical dosimetry was shown to be the very favorable method for radiation field of multi-kilocurie gamma-ray sources, while the Ag-activated phosphate glass needle seemed to have some advantage over the chemical method, especially for isodose mapping of gamma-ray fields.

## INTRODUCTION

The gamma-ray irradiation facility, of which construction is reported by Okamoto, Nakayama and Takahashi in the preceding paper[1] in this Bulletin, was completed in the beginning of 1958, and was opened to many workers in the University for their use from June, 1958. Prior to a program of studies on the effect of gamma-rays on a variety of chemical and biological materials by the use of the facility, it had beed desirable to determine the gamma-ray dose rate inside an irradiation cavity. The authors worked on the dosimetry, reported in this paper, as they were asked to be in charge of the operation and maintenance of the facility.

For the dosimetry of the intense gamma-radiation field, several methods have been proposed by many workers ; chemical dosimetry (ferrous-ferric, ceric-cerous, methylene blue, and nitrous oxide) and physical dosimetry (small ionization chamber, Ag-phosphate glass needle, and adiabatic calorimeter) are used generally for this purpose.

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* 清水 栄, 田中 伸, 中山康之

The chemical dosimetry using the ferrous-ferric reaction has been reported by many workers[2~14] as one of the best methods for high intensity gamma-ray field, accurate, simple to use and giving reproducible results. The dosimeter utilizing Ag-activated phosphate glass has been developed by Schulman and others[15~20] and found to be reliable method with some favorable properties for measuring gamma-ray intensities. In the present work the chemical dosimetry was applied as the main procedure, while Ag-activated phosphate glass needle dosimeters were also used for reference in parallel with the chemical method. Further, the mathematical estimation of dose rates was undertaken to compare those with the experimental results, by assuming many conditions to simplify the calculation.
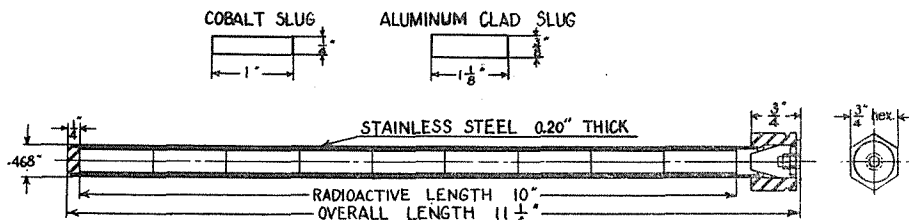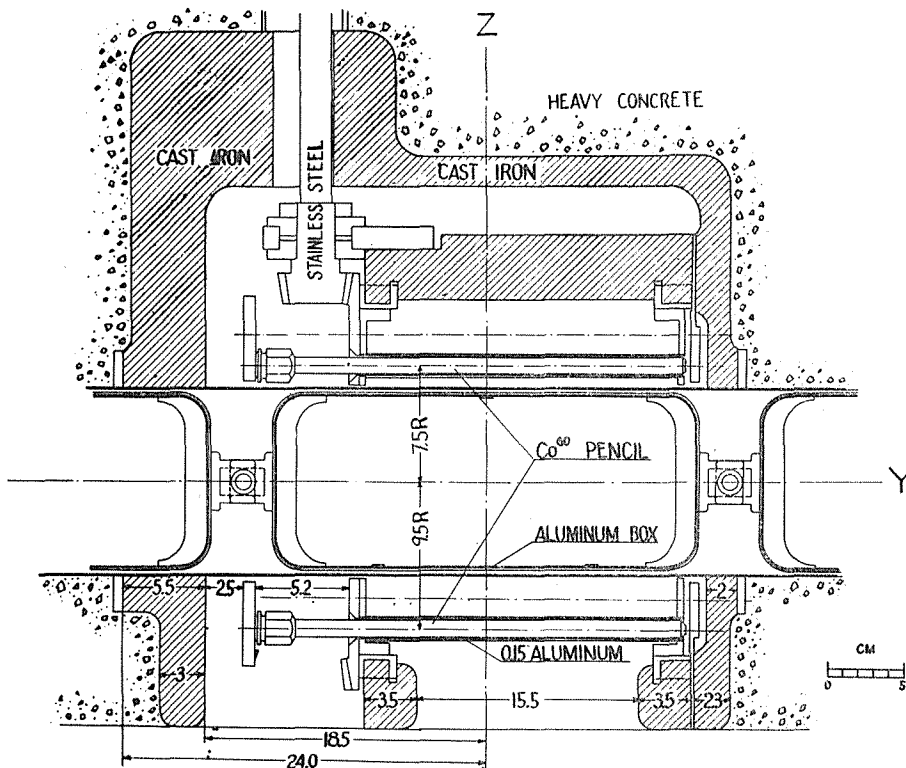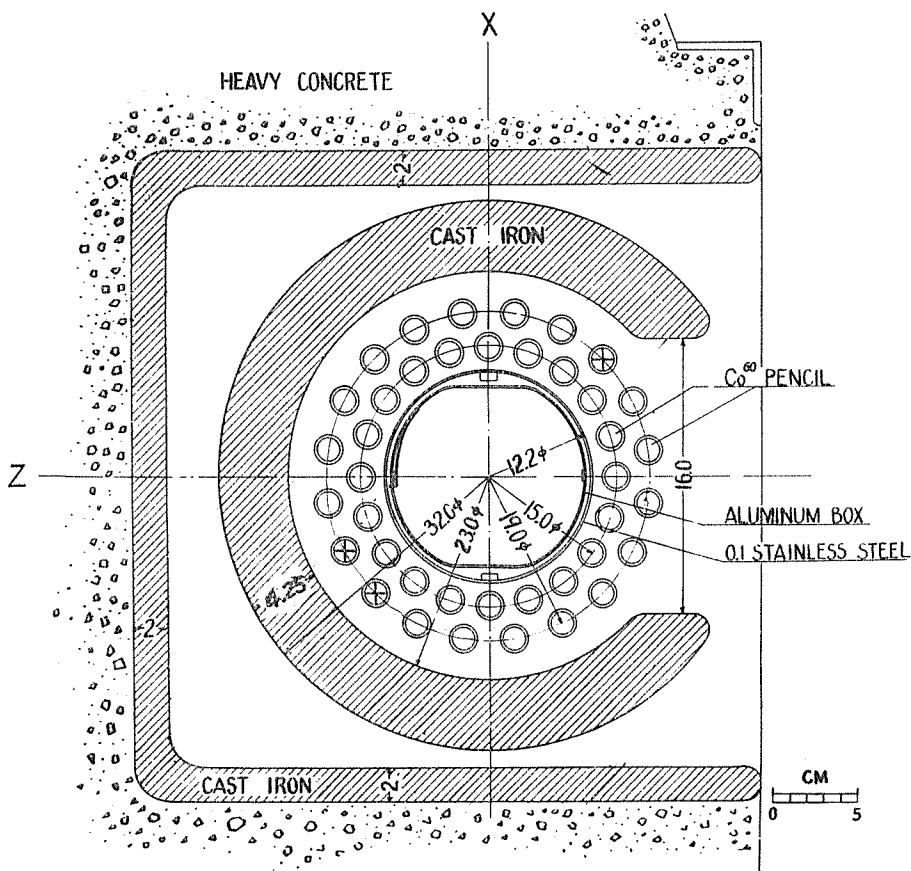
## Co⁶⁰ GAMMA-RAY SOURCE AND IRRADIATION CAVITY



Fig. 1. AECL Co⁶⁰ pencil with a nominal rating of 61.7 Curies (June, 1957).



(a) Horizontal cross section at the center

(b) Vertical cross section at the center

Fig. 2.　Cross sections of the irradiation cavity with 40 Co[60] pencils.

　　　The structure of the present Co[60] irradiation facility is described in detail in the preceding paper[1] of this issue except for numerical details of some parts.　To understand the present work some details of the gamma-ray source and irradiation cavity are desirable to describe.

　　　The source consists of a bundle of 40 Co[60] pencils, each contains 9 cobalt slugs of 1/4 in. in diameter and 1 in. long.　Each slug is encapsulated in thin aluminum jacket and 9 slugs are contained in a thin stainless case, forming a pencil with 10 in. active length, as shown in Fig. 1.　The Co[60] pencils produced by the Atomic Energy Canada Limited at Chalk River were given a nominal rating of 61.7 Curies for each pencil (June, 1957).　The pencils are mounted in a circular holder, whose inside diameter is about 15 cm.　The pencil-holder assembly and irradiation cavity are shown in Fig. 2.　The whole assembly, having about 2100 Curie Co[60] (April, 1958), is embedded in heavy concrete shield as shown in Fig. 2.

　　　For the irradiation experiment, the sample to be irradiated is placed in an aluminum box and then transported into the center of the irradiation cavity by a mechanical conveyer system.　The cavity is the region of interest

for dosage measurements with this source.

## MEASUREMENTS OF DOSE RATE

For our dosimetry the Fricke ferrous sulfate dosimeter was adopted by considering the dose rate in the cavity to be the order of $10^5$ r/hr, as well as from the reason that this famous chemical dosimetry is known to have some good characteristics very favorable for our present purpose; be independent of intensity over a wide range and also independent of wave length, slight temperature changes, be reproducible and easily made from shelf reagents, be accurately measurable, and be simple and convenient to use. Further, it is noted that the responsibility of the Fricke method is known to be linear with respect to dose from 4 kr to 40 kr[6,12].

The Fricke solution used in the present work was made by the following procedure. Two gm of $(NH_4)_2Fe(SO_4)\cdot 6H_2O$, 0.3 gm of NaCl and 110 cm³ of conc. $H_2SO_4$ were added in sufficient distilled water to make 5 liters of solution[12]. Such solution with 0.8N $H_2SO_4$ was irradiated in thin glass ampoules, 18 mm internal diameter and was filled to a depth of about 30 mm. The ampoules of solution were used as dosimeters throughout the work. The dimensions of the glass ampoules were chosen such as those above mentioned by reflecting the fact reported by Weiss[6,12], who showed that the ferrous oxidation rate increased markedly in cylindrical cell as the internal diameter fell below 8 mm; this diameter effect presumably arises from the action of secondary electrons generated from the glass wall. The dosimeters were assembled in the aluminum sample box and transported into the irradiation cavity where dose rate to be measured.

The absorbance at 305 mμ of both irradiated and non-irradiated dosimeter solution was measured in the 1 cm cell by a photoelectric spectrophotometer (Hitachi EDU-2A) with a slit width of 0.3 mm. Ferric ion was determined photometrically at 305mμ[5].

The dose rate $R$ is given by the following expression;

$$R \text{ (r/hr)} = \frac{10^9}{\varepsilon Y} \times \frac{A - A_0}{T}, \tag{1}$$

where $\varepsilon$ is the molar extinction coefficient, $Y$ the ferric sulfate yield in units of micromoles per liter per 1000 r, $A$ and $A_0$ the absorbance of the irradiated and non-irradiated solution, respectively, and $T$ is the time of irradiation in units of hours.

Since the absorbance depends on the temperature of solutions, the measured absorbance were corrected to the value at a standard temperature, 17.5°C. To obtain this correction factor we measured the absorbance vs. temperature relation for 15 ppm ferric solution (0.8 N $H_2SO_4$). The result obtained is shown in Fig. 3, giving a temperature coefficient, +0.0025/°C. From this result it was able to get the relations of absorbance vs. concentration of Fe+++ and temperature coefficient of absorbance vs. concentration of Fe+++, as shown in Figs. 4 and 5, respectively. By this procedure the temperature correction

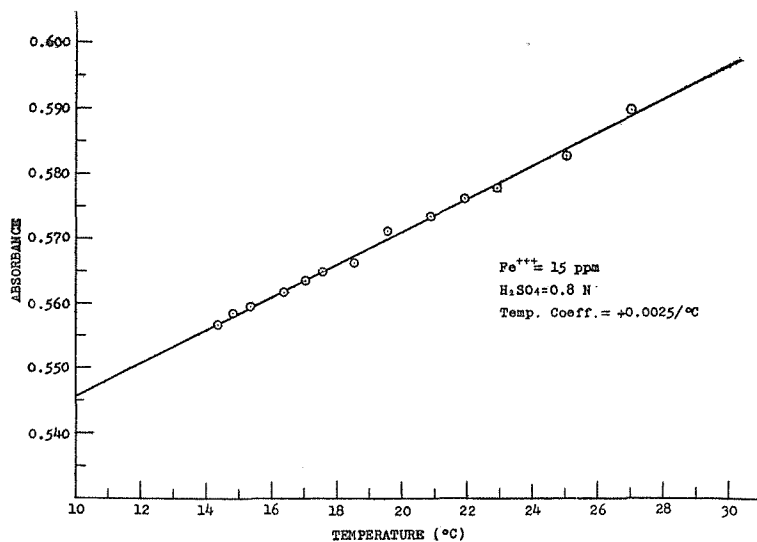Fig. 3. Absorbance *vs.* temperature relation at 304 m$\mu$ for the ferric solution.
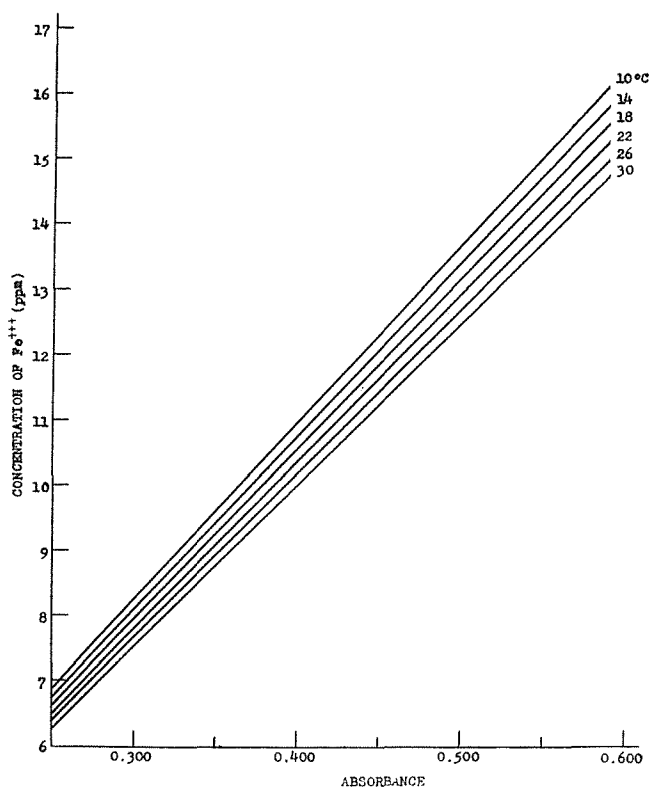
Fig. 4. Absorbance as a function of the concentration of Fe$^{+++}$ in 0.8N H$_2$SO$_4$.
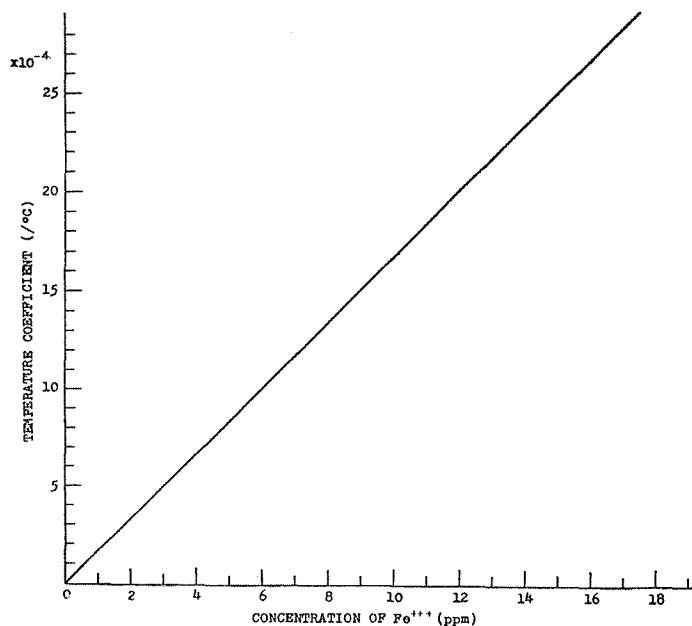
Fig. 5. Temperature coefficient of absorbance as a function of the
concentration of Fe$^{+++}$ in 0.8N H$_2$SO$_4$.

factor was obtained, necessary to normalize the measured absorbance to the
value at 17.5°C.

To obtain the value of the molar extinction coefficient, $\varepsilon$, in Eq. (1), we
observed absorbance of the solutions specially prepared, containing Fe$^{+++}$ of
5, 10, 15 and 20 ppm but with 0.8 N H$_2$SO$_4$ as in the dosimeter solution, and
the value of $\varepsilon$ was determined to be 2059 at 17.5°C, which was in good agree-
ment with the value given by Weiss and others, 2080 at 17.5°C, derived from
their value, 2174, at 23.7°C by using their temperature coefficient[12].

It was somewhat difficult to determine the value of $Y$ in Eq. (1), since in
our University there was no standard Co$^{60}$ gamma-ray source, whose activity
had been measured accurately. Therefore, in order to expose our dosimeter
ampoules in the gamma-ray field with an accurately known intensity, the am-
poules were sent to the National Institute of Genetics at Mishima and were
irradiated up to doses of 5 kr and 10 kr by the use of the Co$^{60}$ facility there.*
Absorbance measurements were then carried out by our spectrophotometer on
these dosimeter solutions exposed to 5 kr and 10 kr. By this procedure the
value of Y was found to be 15.79 $\mu$Mol/1000 r, showing a fairly good agreement
with 16.0±0.3 $\mu$Mol/1000 r given by Weiss and others[12] with the similar Fricke
solution, although our value seemed to have a probable error of several per-
cent due to uncertainties included in the dose estimation.

The time of irradiation for the dosimeters was chosen so as to maintain
a maximum total exposure of less than 20 kr, corresponding to irradiation for

* The authors are indebted to Dr. S. Kondo for his help in this irradiation.

about 2–4 minutes ; in this region of gamma-ray intensities the ferrous-ferric dosimeter was reported to be very reliable by many workers as described in the preceding chapter. By reflecting this condition and the sensitivity characteristic of the spectrophotometer used in the present work[21], and, further, to eliminate the effect of strayed radiation in the neighbouring space outside the cavity, through which the dosimeter is carried into and pulled out, difference of absorbance of the dosimeter solutions irradiated for 4 and 3 minutes at the fixed point in the cavity was measured as a value giving true dose rate free from the undesired effect. By this procedure we could observe the increase of absorbance of the solution by the gamma-ray irradiation of one minute at the fixed point in the cavity.

The experimental arrangement of dosimeters in the cavity and experimental values of the dose rates obtained at the points where the dosimeters were placed are shown in Fig. 6. The $(x, y, z)$-coordinates of centers of dosimeters are given in parentheses in the figure. The origin of the coordinate-system is taken in the center of the cavity, and $x$-, $y$- and $z$-axis are taken as shown in Fig. 2. These measured values shown in the figure are all normalized to April, 1958. Our results are the mean values of those obtained by four independent runs of measurements performed at times separated by an interval of one week from April to May, 1958. The average deviation from the mean value is less than 1.6 percent for all cases. It should be noted that as the dosimeter occupies some spatial extent, cylindrical region of about 18 mm in diameter and 30 mm long, the experimental values obtained give only the spatially averaged values of gamma-ray intensities in the region occupied by the solution ampoules. The size of our dosimeter was too large enough to average out the intensities with different values, especially when the dosimeter was placed near the inner wall of the cavity where intensity gradient is much larger than at the center region.

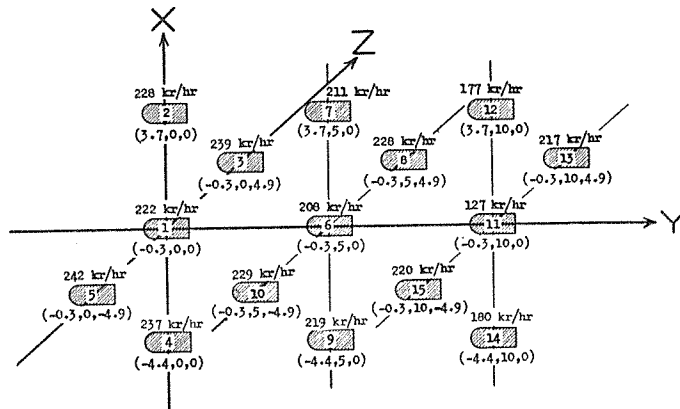The intensity due to strayed radiation at the mid-point of the adjoining



Fig. 6. Showing schematically experimental arrangement of dosimeters and measured dose rates in the cavity. (Normalized to April, 1958). The (x, y, z)-position of centers of dosimeters are shown in units of cm in parentheses.

aluminum box outside the cavity was measured by the dosimeter placed at this position and 13.2 kr/hr (normalized to April, 1958) was obtained.

It was felt rather difficult to apply the ionization chamber to our purpose, since the irradiation cavity of the present facility is embedded deeply in the heavy concrete shield. However, in the middle of November, 1958 the authors were fortunately able to use the Ag-activated phosphate glass needle by the kind assistance of Dr. S. Kondo* at the Biology Division, Oak Ridge National Laboratory, Tennessee. Then this glass needle dosimeter, having advantage over the chemical method for gamma-ray dosimetry in some aspects, was used in our cavity. The small Ag-activated phosphate glass needle, 1 mm in diameter and 6 mm long, were embedded in a Lucite cylindrical shield, as shown in Fig. 7. The Lucite shield with proper thickness was necessary to get secondary electron equilibrium in the glass needle when it was used as a dosimeter to give the firm value of dosage. The needles were placed at the center of the cavity and irradiated to about 2, 4, 6 and 8 kr, respectively. The time of irradiation for each needle was carefully adjusted by considering our experimental value of dose given by the Fricke dosimeter and the estimate of dose which the needle might receive during its transport to and from the point to be measured.
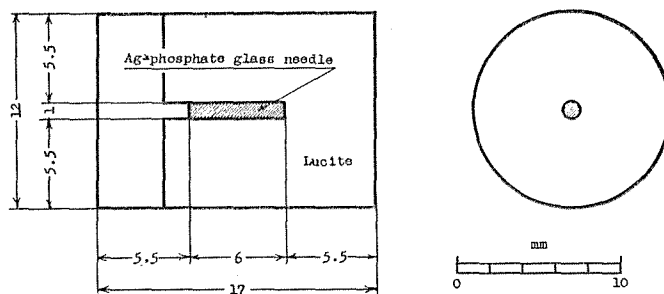


Fig. 7. Ag-phosphate glass needle embedded in Lucite shield.

The glass needles with and without Lucite shield, both irradiated to the assigned doses, were sent back to the Oak Ridge National Laboratory. By measuring the gamma-ray-induced fluorescence of the needles under excitation by ultraviolet light, Dr. Kondo decided the dose rate being 11 percent larger than the value given by our Fricke dosimeter placed at the center of the cavity[22]. The discrepancy between these two dosimetries will be discussed in the following chapter.

It should be noted that throughout the present dose measurements our facility had only 37 Co[60] pencils ; positions of 3 pencils then lacking are shown by the mark "+" in Fig. 2(b).

## CALCULATION OF DOSE RATE

The geometry of the present Co[60] source was so complicated, as described

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* Then on leave from the National Institute of Genetics, Mishima, Shizuoka-ken, Japan.

in the preceding chapter, that exact mathematical treatment involving many actual conditions was very difficult. To simplify the problem and to get an approximate solution we had to use some assumptions.

The actual size and construction of our source pencil is shown in Fig. 1, however, for the present calculation it is assumed that all activities are concentrated on the longitudinal axis and distributed uniformly over this axis. Self-absorption of gamma-rays in a pencil may be approximated to be the absorption by thickness of cobalt surrounding the theoretical axis of concentration of Co[60]. Absorption in a thin stainless steel wall and aluminum case and self-scattering in the pencil were neglected. It was also assumed that the scattering radiation due to the complicated structure of the irradiation cavity does not affect the dose rate to be estimated.
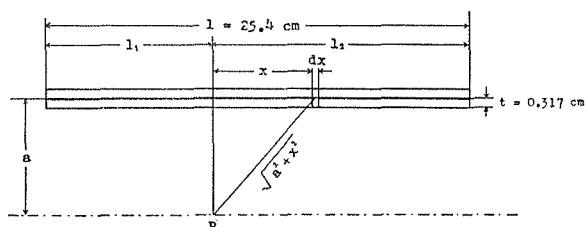


Fig. 8. Geometrg of a pencil source having the activity on its longtudinal axis.

At first, we calculated the intensity at any point inside the cavity contributed by one source pencil. Then the contributions from each rod calculated by simiar procedure were summed up and the total dose rate at a point inside the cylindrical array of source pencils was found. On this assumed Co[60] pencil, as shown in Fig. 8, the intensity at a point P from an element of length $dx$ is given by:

$$dI = \frac{Sadx}{a^2 + x^2} \exp\{-\mu t(a^2 + x^2)^{1/2}/a\}, \tag{2}$$

where

$S = $ r/hr per 1 C Co[60] at 1 cm $= 13.56 \times 10^3$ r/hr/C at 1 cm,

$a = $ linear specific activity $= 2.18$ C (normalized to April, 1958),

$l = l_1 + l_2 = $ active length of a source pencil $= 25.4$ cm,

$t = $ active radius of a source pencil $= 0.317$ cm,

$\mu = $ linear absorption coefficient of Co[60] gamma-rays for cobalt $= 0.447$ cm$^{-1}$.[23]

$S$ was calculated by the following expression:

$$S = \frac{(3.7 \times 10^{10} \times 3600)\{(1.17 \times 3.53 \times 10^{-5}) + (1.33 \times 3.41 \times 10^{-5})\}}{4\pi(6.77 \times 10^4)}$$

$$= 13.56 \times 10^3 \text{ r/hr/C at 1 cm,}$$

where $1/4\pi$ is the fraction of energy emitted by the source that passes through an area 1 cm$^2$ at 1 cm from the source, $(3.7 \times 10^{10} \times 3600)$ is the disintegration per hour per Curie, 1.17 and 1.33 MeV are the gamma photon energies from Co[60], $3.53 \times 10^{-5}$ cm$^{-1}$ and $3.41 \times 10^{-5}$ cm$^{-1}$ are their respective Compton absorption coefficients in air at 0°C and 760 mm Hg, and absorption of $6.77 \times 10^4$ MeV is

equivalent to one Roentgen[24,25,26]. Photoelectric and pair production absorption are negligible for these energies in air.

The total intensity at a given point P is obtained by integrating dI, given by Eq. (2), over the whole pencil as :

$$I_p = Sa \int_{-l_1}^{+l_2} \frac{\exp\{-\mu t(a^2+x^2)^{1/2}/a\}}{a^2+x^2}\, dx.$$ (3)

Expanding in the series $e^{-x} = 1 - x + x^2/2! - x^3/3! + \cdots\cdots$,

$$I_p = Sa \sum_{n=0}^{\infty} \left\{ \left( \frac{-\mu t}{a} \right)^n \frac{1}{n!} \int_{-l_1}^{+l_2} (a^2+x^2)^{n-2/2}\, dx \right\}, \quad n = +\text{integers}.$$ (4)

When $\mu t/a \ll 1$ the expression converges very rapidly, so that terms beyond the third are insignificant. Integrating the first three terms the expression becomes :

$$I_p = \frac{Sa}{a} \left\{ \left( \tan^{-1} \frac{l_2}{a} + \tan^{-1} \frac{l_1}{a} \right) - \mu t \log \frac{l_2 + (a^2+l_2^2)^{1/2}}{-l_1 + (a^2+l_1^2)^{1/2}} + \frac{(\mu t)^2 l}{2a} \right\}.$$ (5)

By this expression the gamma-ray intensity contributed by one Co[60] pencil at a point in the cavity is calculated, and summing up the contributions of 40 pencils the total intensity at the point can be obtained. In the present calculation, however, absorption and scattering of the gamma-rays from the outer array of source pencils at the inner array of pencils were neglected.

By this calculation basing upon several assumptions as above mentioned, the values given in Table 1 were obtained.

Table 1. Calculated dose rates at points on the $yz$-plane in the cavity with 40 pencils. (Normalized to April, 1958, and in units of kr/hr)

| $z$ | $y$ | | | | | | |
|---|---|---|---|---|---|---|---|
| | 0.0 cm | 2.0 cm | 4.0 cm | 6.0 cm | 8.0 cm | 10.0 cm | 12.0 cm |
| 0.0 cm | 235.0 | 232.4 | 227.1 | 216.2 | 200.8 | 179.5 | 153.8 |
| 1.5 | 238.4 | 236.4 | 230.0 | 219.9 | 204.3 | 182.3 | 155.8 |
| 3.0 | 248.7 | 246.4 | 240.9 | 230.4 | 214.1 | 191.1 | 161.7 |
| 4.5 | 268.8 | 266.9 | 261.5 | 252.0 | 234.5 | 209.0 | 173.9 |
| 6.0 | 312.0 | 309.5 | 303.7 | 293.9 | 276.5 | 250.9 | 203.0 |

In Fig. 9 calculated isodose curves on the $yz$-plane are mapped. As shown in Fig. 2(b), $x$- and $y$-axis pass through centers of pencils of the inner array, so that if these axes are taken so as to pass through the centers of pencils of the outer array, i.e. $xy$-coordinates incline 9° to our system, isodose curves on this $yz$-plane are expected to have values very slightly smaller than those given in Fig. 9. However, calculations showed differences are less than 0.1 percent. So that it may be reasonable to say that isodose surfaces in the cavity can be given by surfaces of revolution of the isodose curves in Fig. 9 around the $y$-axis.
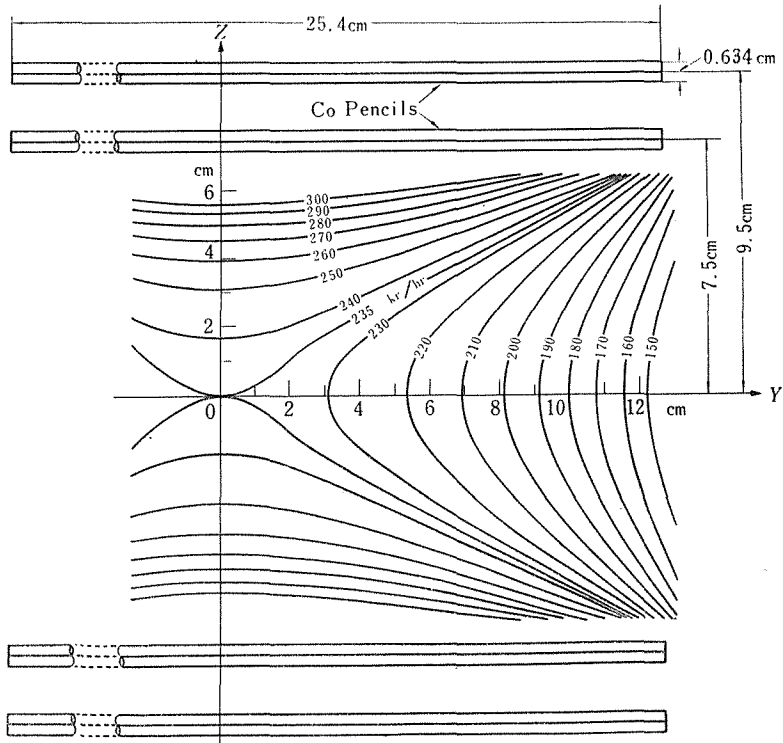
Fig. 9. Calculated isodose curves on the yz-plane with 40 Co[60] pencils. (Normalized to April, 1958)
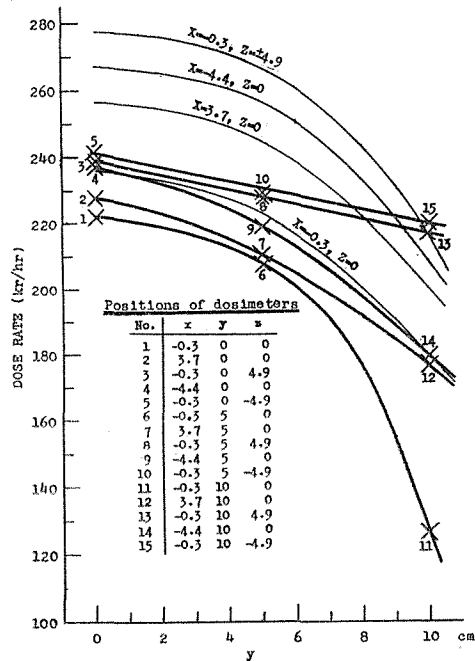
Fig. 10. Measured and calculated dose rates along the lines parallel to the y-axis. (Normalized to April, 1958).

## DISCUSSION

The measured and calculated values of dose rates are shown in Fig. 10. The thick curves are experimental, each of which is drawn so that it joins values observed by three different chemical dosimeters arranged along the line parallel to the *y*-axis. The positions of dosimeters are also given in the table in the figure. The calculated dose rates corresponding to the experimental curves are shown by thin curves. However, it is noted that calculations were performed with 40 source pencils while the measurements were made with only 37 pencils as discribed in the preceding chapters. For 37 pencils (1940 Curies, April, 1958) used actually in the measurement the calculated dose rate at the mid-point of the cavity was obtained as 219.5 kr/hr. The value given by the No. 11 dosimeter showed the smaller value than that expected may be due to the effect of aluminum reinforces attached to the end of the aluminum box.

It is of interest to give few accounts on the comparisons between observed and calculated values for the present gamma-ray field, although such discussion is not so meaningful, since our calculations were performed basing upon many assumptions differ from the actual conditions and, further, no firm estimate of the activities of the $Co^{60}$ sources were supplied by the maker. The calculated and experimental values at the center of the cavity obtained by different methods are summarized in Table 2. The ratios of measured-to-

Table 2. Dose rates at the center of the irradiation cavity with 37 pencils by measurements and calculations (Normalized to April, 1958).

| Method | Dose rate (kr/hr) | Ratio=$\frac{exp.}{cal.}$ |
|---|---|---|
| Ferrous-ferric dosimeter | 222 | 1.01 |
| Ag-phosphate glass needle | 246 | 1.12 |
| Calculation | 219.5 | |

calculated dose rates appear in the right column. The ratio of the values by two different measurments is 1.11. This discrepancy between two experimental values is presumably due to the fact that the value observed by the Fricke dosimeter is the spatially averaged value over the region occupied by the dosimeter solution while the glass needle having smaller dimensions gives the value almost corresponding to that at the point to be measured. However, since our both experimental values seem to have probable errors of about ±5 percent, the agreement within 11 precent between these two measured values may be said to be considerably good. The effect of strayed gamma-rays was neglected in our calculation basing upon many assumptions may explain the discrepancy between measured and calculated values. Considering many conditions above discussed these values by different methods are in reasonably good agreement, and the dose rate just at the midpoint of the cavity is con-

Sakae Shimizu, Shin Tanaka and Yasuyuki Nakayama

cluded to be 234 kr/hr ±5 percent.

From the present work it may be concluded that for the rough estimation of intensities of the gamma-ray field the ferrous-ferric dosimeter exhibits to be very useful while for the detailed mapping of isodose curves or surfaces the Ag-activated phosphate glass neelde has some advantage over the chemical dosimetry.

## ACKNOWLEDGMENTS

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