A Cobalt-60 Irradiation Facility for Radiation Chemistry

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A gamma-ray irradiation facility with about 2000 Curies of Co^{60} had been installed at the Takarazuka Radiation Laboratory of the Sumitomo Atomic Industries Ltd. in August, 1959. Some consideration was paid on its design and construction. Some details of design and construction of the facility are described. Dosimetry for the gamma-ray field where samples to be irradiated was carried out by the use of the ferrous-ferric chemical method and by a Victoreen r-meter of Radocon type. The theoretical estimation of dose rates was also performed by adopting some assumptions on configuration and arrangement of sources. The comparison between both estimations of dose rates, experimental and calculated, is· discussed.

INTRODUCTION

Since there is an increasing interest in radiation effect on a variety of materials in industrial field, a Co^{60} irradiation facility as a research instrument with special features for radiation chemistry was installed at the Takarazuka Radiation Laboratory as one of research projects of the Sumitomo Atomic Industries Ltd. in August of 1959. The facility with 2000 Curies of Co^{60} was housed in a shielded corner of the main building of the Laboratory. The total unit was constructed by design principles that achieved the maximum exposure rate to be about 1×10^6 r/hr. This radiation dose rate·is delivered by about 2000 Curies of Co^{60}, however,·our facility is designed to be able to store about 5000 Curies without any improvement of construction. The facility is already in use for general irradiation researches, especially for studies on irradiation effects of some chemical materials of industrial interests. In the present paper some features of construction of the facility are given and dose rate estimation by experimental as well as theoretical procedures is also reported.

CONSTRUCTION

Source. The source cage made of stainless steel holds up 140 pieces of Co^{60} source of the coin type in 10 hollow pencils in vertical position, as shown in Fig. 1.
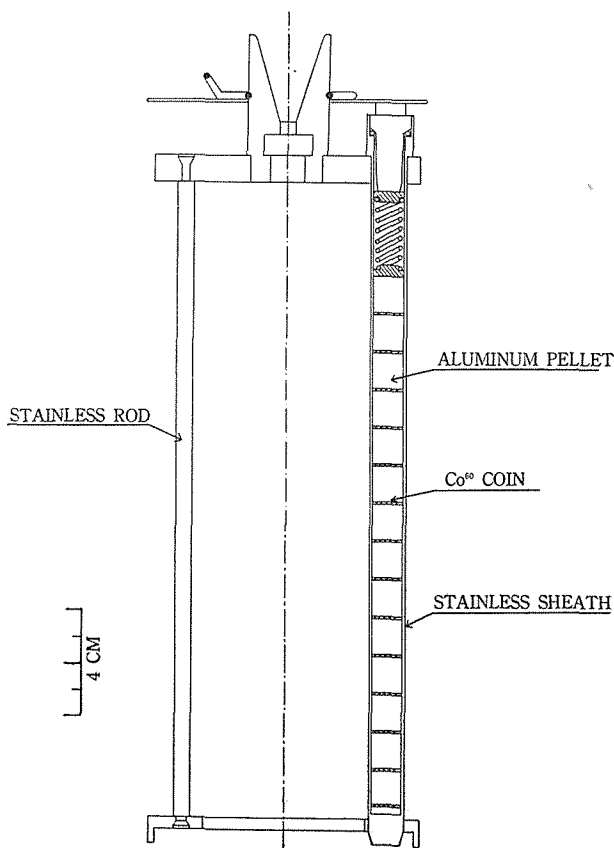
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Fig. 1. Vertical section of the source cage.

Fourteen pieces of Co^{60} coins, spaced each other by an aluminum pellet, are inserted into each pencil with an inside diameter of 10.1 mm, a wall thickness of 1 mm, and an effective length of about 230 mm, as shown in the figure. Each coin of 10 mmϕ $\times 1$ mm thick with the nominal rating of about 13.7 Curies (August 19, 1959) was supplied from Oak Ridge National Laboratory. The total strength of the source is, therefore, about 1925 Curies. The whole source assembly is surrounded by a stainless steel sheet of 1 mm thick.

The annular source geometry provides a symmetrical gamma-ray field inside and outside the source assembly. The sample being irradiated outside the source can be rotated, if necessary, to get the uniform effect of irradiation on the sample. Stainless steel is used for the source cage since corrosion is promoted by the acid nitrogen oxides and O_3 produced in the irradiated air.

Mechanical. To insure the safe loading of materials to be irradiated on the sample table, the whole source assembly can be lifted into the cave located at the ceilling through the protecting cylinder of stainless steel by a motor-driven mechanism which is operated in a separated control room. By a special electrical and mechanical device anybody can enter into the irradiation room only when the source assembly is lifted into the ceiling and the thick lead shutter closes so as to keep the source perfectly in the cave. When the sample loading is completed and

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a worker escapes from the irradiation room and closes its door, an operater can move down the source cage onto the regular position for irradiation.

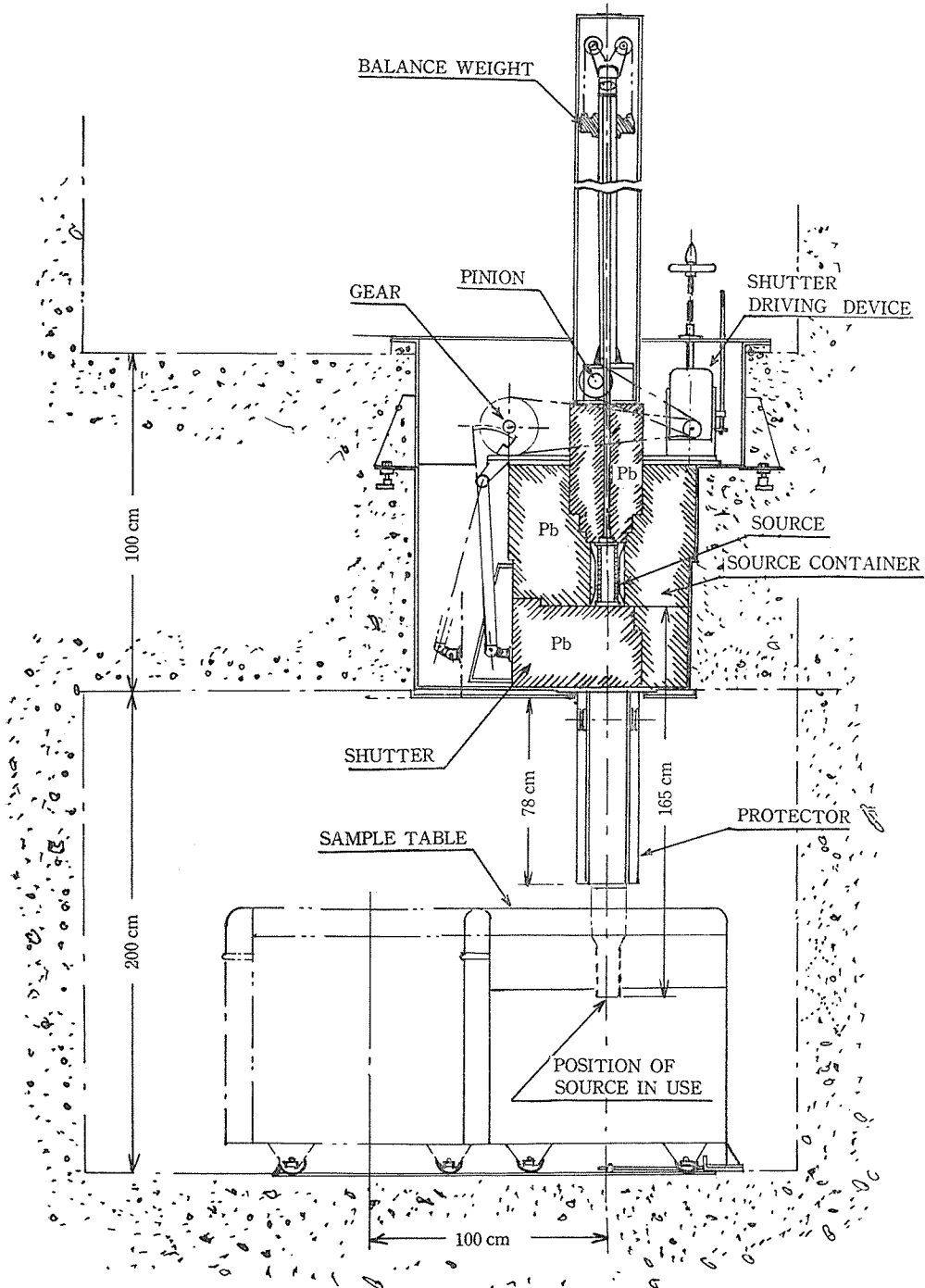The construction of the facility, including the cave, drive mechanism and
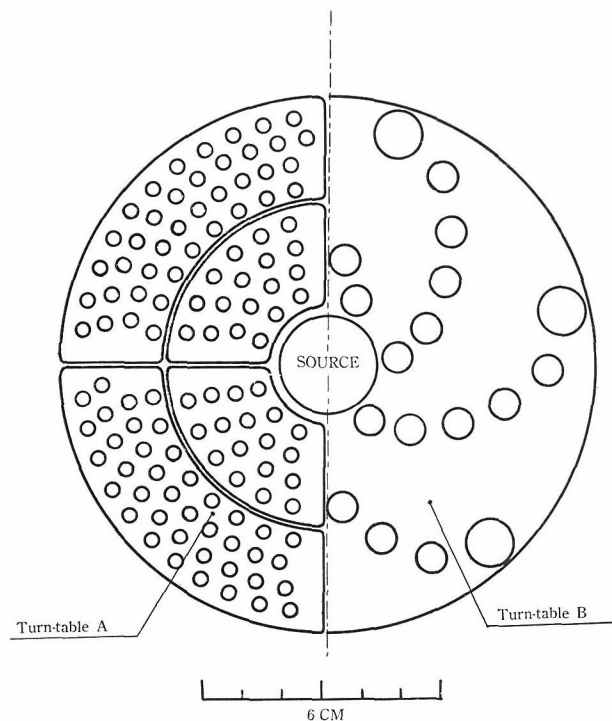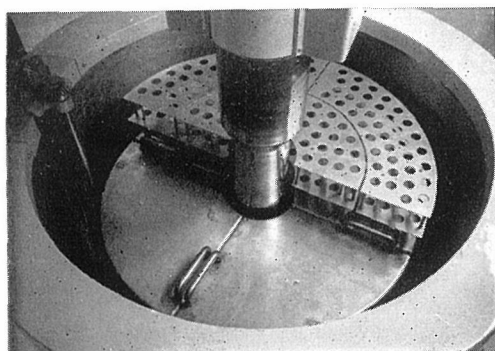

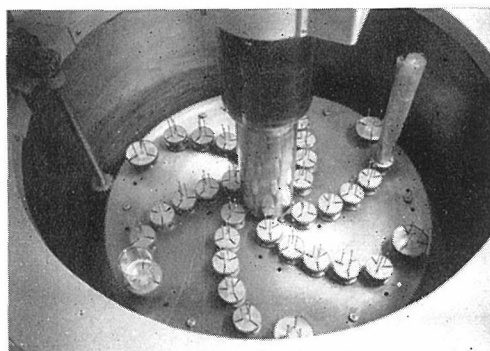Fig. 2. Elevation view of the irradiation facility.

Fig. 3. Plan view of the irradiation turn-table. Small circles show the holes perforated into the aluminum turn-table, where test tubes or small vessels containing materials to be irradiated are placed.

(a)　Turn-table **A**.　　　　　　　　(b)　Turn-table B.

Fig. 4.　Irradiation turn-tables.

irradiation table, is shown in Fig. 2. While, Figs. 3 and 4 show the details of irradiation turn-table, which revolves horizontally around the center axis once per minute. We have two kinds of such turn-tables, easily changeable each other by the need of research. One can hold 232 test tubes of samples, and the other can hold 36 small vessels of about 4 cm and 6 cm in diameter on the small turn discs being rotated on its axis slowly, one turn per minute (see Fig.4). By means of this device uniform irradiation of samples is achieved.
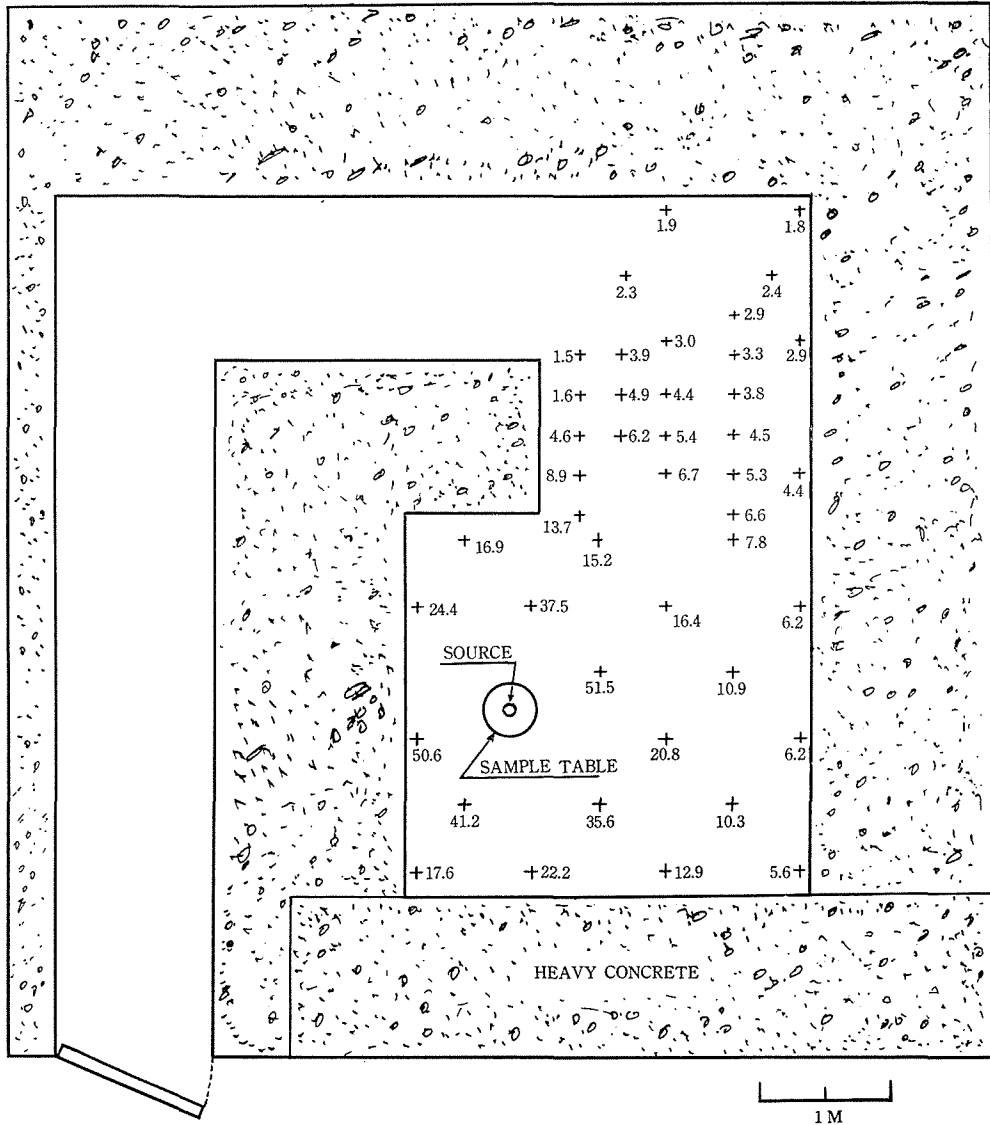
Fig. 5. Plan view of the irradiation room. Intensities of stray radiation
are in units of r/min, measured by a Victoreen r-meter (Model 575 A
Radocon) in the end of August, 1960.

Irradiation room. Fig. 5 shows the plan view of the irradiation room, showing
also observed data of intensities of stray radiation in units of r/min when the source
cage is at the operating position. These values were measured by a Victoreen
r-meter (Model 525A Radocon) in the end of August, 1960.

MEASUREMENTS OF DOSE RATE

Dose rates were measured chemically by the ferrous-ferric dosimeters. The
principle of this dosimetry bases upon the production of ferric ions by the irradi-

ation of gamma-rays in the dilute solution of ferrous sulfate in about 0.8 N sulfuric acid solution for dose, in general, of between 5 and 40 kr[1,2]. For quantitative determination of the ferric ions produced, the spectrophotometric method is generally used, which makes use of a spectral absorption peak of ferric ion at about 305mμ in the ultra-violet region. The responsibility of this chemical dosimetry is, however, reported by Danno et al.[3] to be reliable in the range of dose from 4 to about 40 kr for air saturated solution, and also to about 200 kr for oxygen saturated and 3000 kr for nitrogen saturated.

In the present work we used two different solutions: (1) N_2-saturated one for dosimetry inside the source cage, (2) O_2-saturated one for the field outside the source cage.

For purification of water, which was used throughout the work, we adopted the method reported by Johnson and Weiss[4]. Several reagents used are, of cource, chemically special grade of purity. Dilute solutions of $FeSO_4 \cdot 7H_2O$ ($4 \times 10^{-3} Mol$) and NaCl (1×10^{-3} Mol) in 0.8 N H_2SO_4 solution saturated by O_2 were used for the external field, while the solutions of same components but with 10 times larger quantity of $FeSO_4 \cdot 7H_2O$ and saturated by N_2 were used for the internal field of the source cage, reflecting on the reliability of these solutions for the range of gamma-ray intensity above mentioned. The solutions of about 12 cc were irradiated in glass test tubes, as shown in Fig. 6. The solutions in these tubes were irradiated on the turn discs on the sample turn-table. The exposure time was chosen to be just one hour, since it seems to fall within the range of this chemical method.

In converting the chemical yield of ferric ions to radiation dosage a value of 8.5 μM/l/kr for N_2-saturated solutions and that of 16.1 μM/l/kr for O_2-saturated ones were used. The molar extinction coefficient was measured by observing absorbance of the standard solutions specially prepared for this object, and we got a value of 2174 at 25°C.
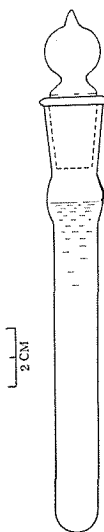

Fig. 6.   Glass test tube of the ferrous-ferric dosimetry.

In physical determination of dose rates a Victoreen r-meter of Radocon type (Model 575A) was employed. In order to compare experimental values obtained by these chemical and physical dosimetry, we measured at the same points of gamma-ray field; the center of the probe of the r-meter was carefully adjusted at the points of centers of chemical dosimeter tubes. For the dose rate higher than 5×10^4 r/hr a probe of No. 603 type and for the field strength less than this value a probe of No. 607 were used.

The experimental dose rates outside the cage obtained by these both methods (the end of August, 1960) are shown in Figs. 7 and 8. Although it is known that both methods adopted in the present work have inherently experimental errors from about 5 to 10 percent, it is interesting to find a fairly good agreement between the measured values obtained by two different procedures. Some difference between both measurements in Fig. 8 is probably due to small displacements of the positions where the chemical dosimeter and probe of the r-meter used were placed.

The dose rates measured by the N_2-saturated chemical dosimeter at the points along the central axis of the source cage, 5 cm and 10 cm from the effective bottom surface, are 5.34×10^5 and 5.45×10^5 r/hr, respectively.
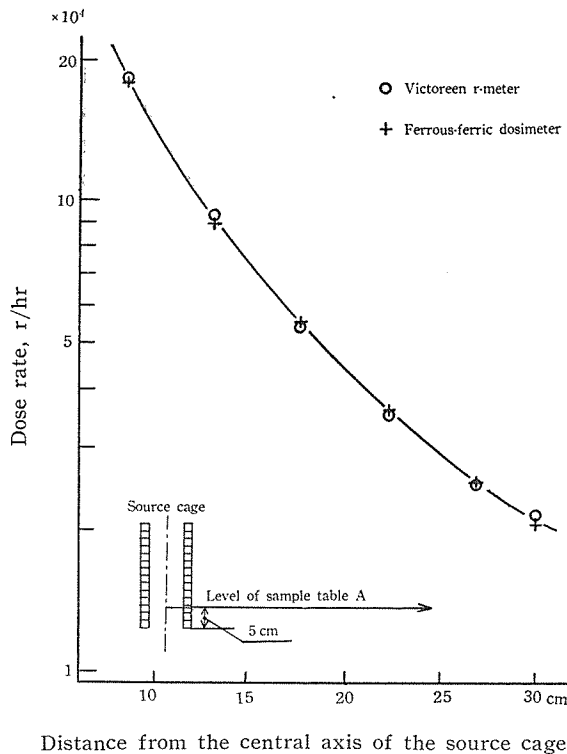

Distance from the central axis of the source cage

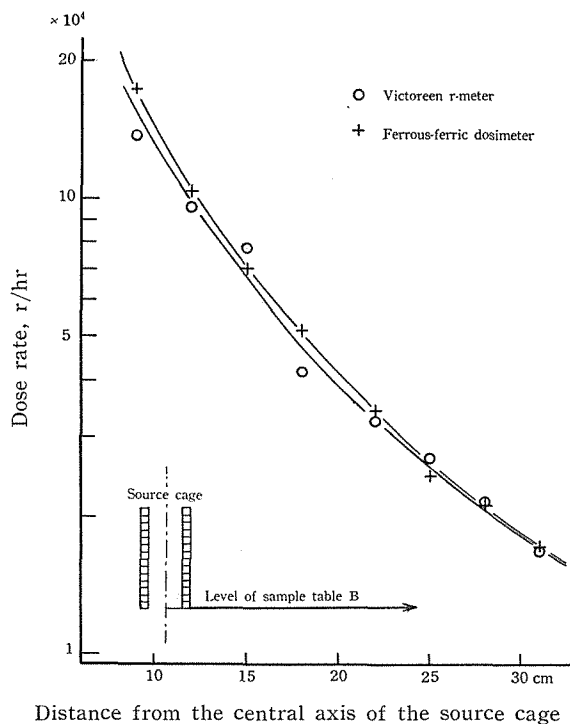Fig. 7. Experimental dose rates. Dosimeters were placed on the sample table A. (End of August, 1960)

Fig. 8. Experimental dose rates. Dosimeters were placed
on the sample table B. (End of August, 1960)

CALCULATIONS OF DOSE RATE

Prior to the extensive use of the facility, in addition to the experimental estimation of dose rates, it is desirable to determine mathematically the amount of radiation to which the samples are exposed at their position. However, since the feature of the Co^{60} source was complicated, it was too much for us to calculate the dose rate exactly. Then, to obtain the approxmiate solution a simplified geometrical arrangement of the source assembly was assumed.

The adopted model is as followings. All activities of a pencil are concentrated on its symmetric axis with uniform specific activity. The gamma photons from this assumed source are self-absorbed in its own pencil, while the absorption by the other materials, aluminum spacers and stainless steel sheath and sheet, is neglected. In the present mathematical treatment, the scattering process is not considered at all. To obtain the dose rate at a given point, the contribution from each pencil to the point concerned was estimated, and then these values are all summed up. If there are other sources on the path of gamma photon, absorption by these cobalt pencils are considered. The effective paths of photons in these nuisances were graphically estimated.

The dose rate at a given point P owing to a dx-part of a pencil is given by the following expression[5] (see Fig. 9.) :

$$dI = \frac{S\varepsilon}{a^2 + x^2} \exp\{-\mu t (a^2 + x^2)^{1/2}/a\} dx, \tag{1}$$
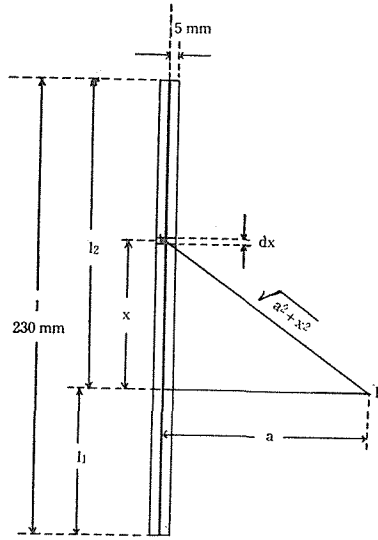
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Fig. 9. Geometry of an assumed pencil source.

where

$S =$ r/hr par 1 C Co^{60} at 1 cm $= 13.56 \times 10^3$ r/hr/C at 1 cm,

$\varepsilon =$ linear specific activity $= 8.37$ C (on August 19, 1959),

$l = l_1 + l_2 =$ active length of a source pencil $= 23$ cm,

$t =$ effective path of a photon in the source and other nuisances,

$\mu =$ linear absorption coefficient of Co^{60} gamma-rays for cobalt $= 0.447$ cm^{-1} [6].

Integrating Eq. (1), we get the approximate expression for one pencil,

$$I_P = \frac{S\varepsilon}{a}\left\{\left(\tan^{-1}\frac{l_2}{a} + \tan^{-1}\frac{l_1}{a}\right) - \mu t \log\frac{l_2 + (a^2 + l_2^2)^{1/2}}{-l_1 + (a^2 + l_1^2)^{1/2}} + \frac{(\mu t)^2 l}{2a}\right\}. \quad (2)$$

The form of the source cage is not of complete cylinder, but composed of ten pencils as mentioned in the preceding chapter. Therefore, the dose rate is not only the function of distance from the cage, but also depends on the direction. Considering the symmetrical geometry we calculated the dose rate on only five planes perpendicular to the central axis of the source cage. ($l_2 = 11.50$, 14.38, 17.25, 20.13,
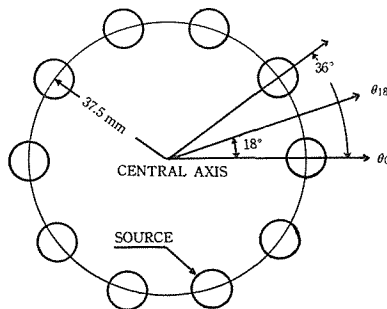

Fig. 10. Diagram showing the directions θ_0 and θ_{18}, in which theoretical values of dose rates were calculated.

and 23.00 cm). And on each plane only two lines are considered, along which dose rates were calculated ; the direction of a line, θ_0, and of the other, θ_{18}, was taken as shown in Fig. 10.

We calculated the dose rate at 12 points with different distances on each line on five horizontal planes, 0, 1, 2, 3, 9, 12, 15, 18, 22, 25, 28 and 31 cm from the central

Table 1. Calculated dose rates in units of kr/hr.
(Normalized to August 19, 1959)

Direction : θ_0

Plane	Distance from the central axis of the source cage (cm)											
	0	1	2	3	9	12	15	18	22	25	28	31
$l_2 = 11.50$ cm	699	715	781	1047	230	142	95.5	68.4	46.9	36.8	29.5	24.2
14.38	690	707	772	1043	225	139	93.9	67.4	46.4	36.4	29.3	24.1
17.25	657	674	741	1016	212	131	88.9	64.4	44.8	35.4	28.5	23.5
20.13	574	591	661	945	186	118	81.4	59.7	42.3	33.7	27.4	22.8
23.00	399	409	447	624	149	100	71.8	55.3	39.1	31.6	25.9	21.7

Direction : θ_{18}

| Plane	Distance from the central axis of the source cage (cm)											
	0	1	2	3	9	12	15	18	22	25	28	31
$l_2 = 11.50$ cm	699	716	779	928	210	134	91.2	66.1	45.9	36.3	29.1	24.0
14.38	690	707	771	904	208	131	89.5	65.1	45.4	35.9	28.9	23.9
17.25	657	674	740	876	196	124	85.4	62.4	43.8	34.9	28.2	23.7
20.13	574	591	657	801	174	111	77.8	57.3	41.5	32.8	27.2	22.6
23.00	399	409	446	529	143	95.8	69.0	52.6	38.3	31.4	25.7	21.6

Distance from central axis of source cage
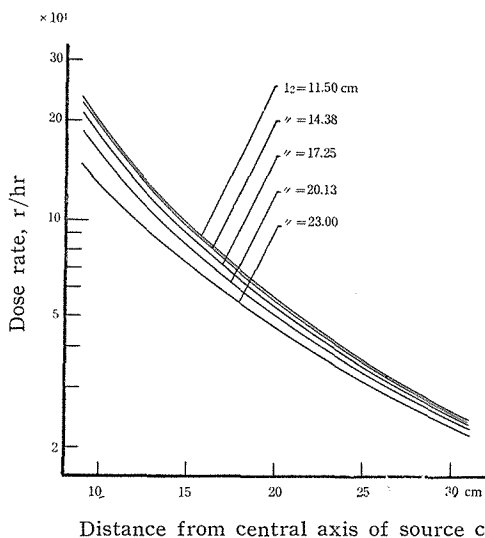
Fig. 11. Calculated dose rate outside the source cage on planes perpendicular to the central axis : Direction θ_0.
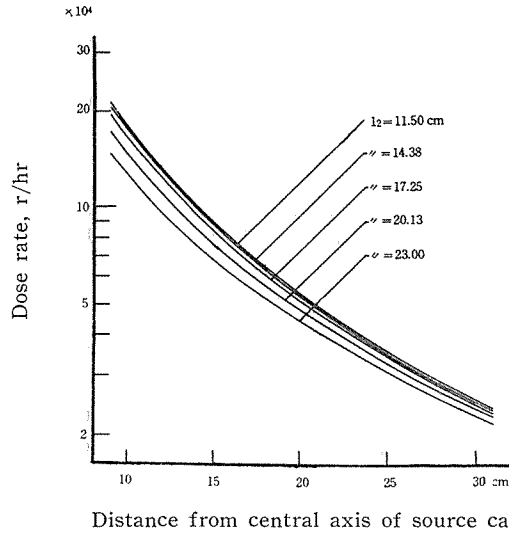
Fig. 12. Calculated dose rate outside the source cage on planes perpendicular to the central axis : Direction θ_{18}.

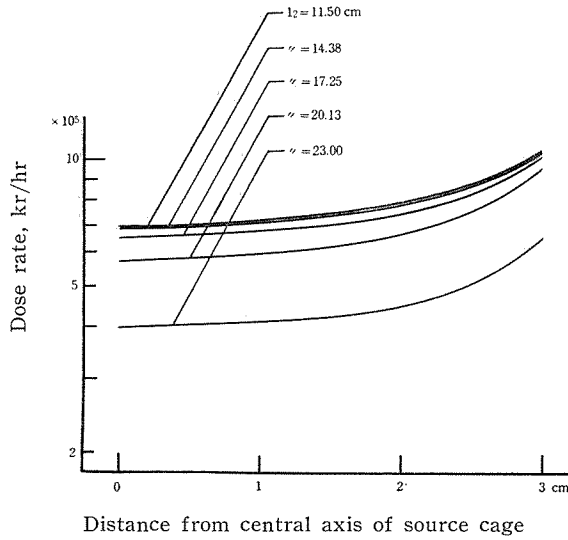Fig. 13. Calculated dose rate inside the source cage on planes perpendicular to the central axis : Direction θ_0.

axis. The results of our calculation are presented in Table 1, and graphically in Figs. 11, 12, 13 and 14. Isodose curves obtained theoretically are also given in Figs. 15 and 16. All calculated values are normalized to August 19, 1959.

DISCUSSION

The measured values of dose rates outside the source cage, shown in Figs. 7 and 8, were averaged ones in direction θ and only refered to the distance from a
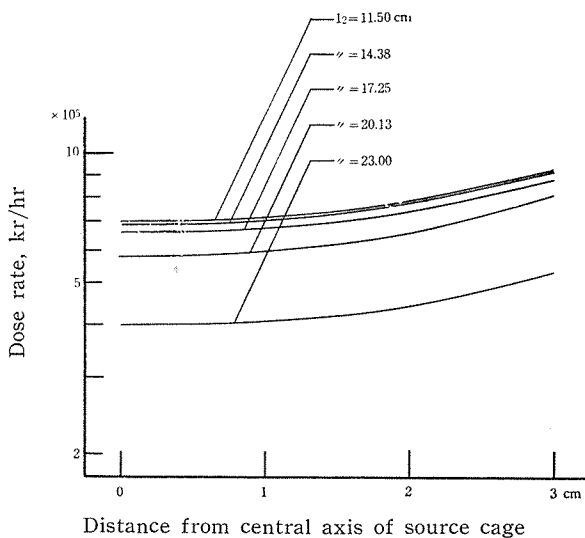
Fig. 14. Calculated dose rate inside the source cage on planes perpendicular to the central axis : Direction θ_{18}.
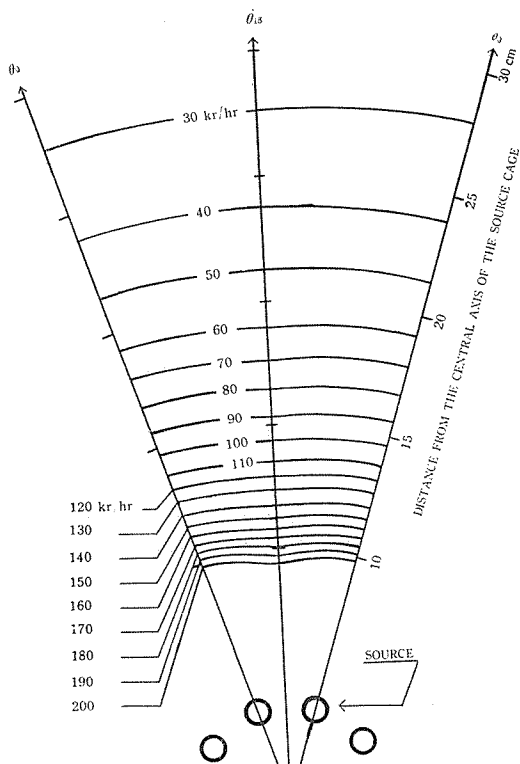

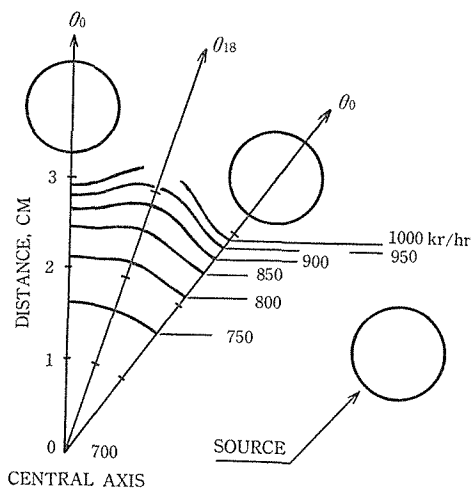Fig. 15. Calculated isodose curves on a horizontal plane $l_2 = 11.50$ cm outside the source cage.

Fig. 16. Calculated isodose curves on a horizontal plane
$l_2 = 11.50$ cm inside the source cage.

central axis of the source cage. The discrepancy between experimental and theoretical values was found to be less than about 10 percent. The comparison of estimated intensities of gamma-ray field by these both methods on the central axis inside the source assembly is given in Table 2. We can not explain properly the discrepancy between both estimations, experimental and theoretical, but some of reasons for it may be due to the fact that our calculation is based upon some assumptions as mentioned in the preceding chapter and the measured values obtained are giving only the spatially averaged values of gamma-ray intensities in the region occupied by the test tube of chemical dosimeter solution or the ionization probe of the r-meter used. However, reflecting many conditions of the present work it may be said that the values of dose rates obtained by both methods are in a fairly good agreement.

For dosimetry of the irradiation domain of such a gamma-ray facility it is desirable to adopt a more elaborate dosimeter with much smaller size and reliable sensibility to large intensities of gamma-ray field, because in such a facility for irradiation purpose the intensity gradient of the field to be measured is, in general, very large.

Table 2. Dose rates on the central axis of the source cage.
(Normalized to the end of August, 1960).

Position on the axis	Method	Dose rate (kr/hr)	Ratio $= \dfrac{\text{exp.}}{\text{cal.}}$
$l_2 = 18.00$ cm	Ferrous-ferric dosimeter	534	1.00
	Victoreen r-meter	522	0.97_8
	Calculation	534	
$l_2 = 13.00$ cm	Ferrous-ferric dosimeter	545	0.95_1
	Victoreen r-meter	555	0.96_9
	Calculation	573	

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