特別掲載

Intercomparison of High Energy Electrons and X-rays in Japan by Use of Fricke Dosimeter

by

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Fricke 線量計による国内高エネルギー電子線およびX線線量の相互比較測定

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国内の37医療施設における高エネルギー電子線およびX線線量の相互比較測定を Fricke 線量計を用いて行つた.

照射によつて生成する第2鉄イオンの定量は，第2鉄とチオシアネートイオンの反応による着色を利用して行つた．この方法は304mμ および225 mμ の紫外吸収による定量法に比べ低線量の測定が可能であつた．測定精度は紫外吸収による方法に比べ，わずかに悪く 1.5%以内であつた．照射容器としては，組織等価なプラスチック容器が望ましいが，線量計溶液をポリスチレン等のプラスチックに保存すると，線量計の感度が変化するので，ガラス容器を用いた．ガラス容器では線量計の感度変化もなく，一定線量照射による化学収率もポリスチレンとの差は認められなかつた.

Fricke 線量計で得られた線量に対する参加施設の線量の比の平均として，電子線で1.01±0.10，X線で1.01±0.09という値が得られ，電子線，X線共に施設間の線量によい一致がみられた.

Introduction

The increasing use of high energy electrons and X-rays in radiation therapy has brought about a need to develop theoretical basis and practical dosimeters for measuring these radiations. Accurate dosimetry is an important factor in determining the optimum single dose and time-dose relationship in radiation therapy. Comparison of results in radiation therapy would have little significance unless the same dosimetric units is employed.

The calorimetric method has been developed as absolute laboratory standard for calibration of dose. However, it is difficult for most medical institutions to conduct such calibration by themselves. Inter-

comparisons, though do not providing doses in absolute units, it may possibly be the best method to obtain uniformity of dosimetry among institutions.

In Japan, Hashizume et al. have reported on the intercomparison of Co-60 γ-rays by fluoroglass dosimeter and that of electrons by LiF thermoluminescence dosimeter[1)2)]. IAEA has made the intercomparison of γ-rays and electrons in chiefly advanced countries using Fricke and LiF dosimeters[3)4)]. The results have been reported in other countries[4)5)]. Fricke and LiF dosimeters have been mainly used in these comparisons because of their high accuracy and relative ease in manipulation. According to Loevinger et al[3)]., higher accuracy can be obtained by Fricke dosimeter than LiF dosimeter when the number of participating institutions is rather small.

The present paper deals with the intercomparison of high energy electrons and X-rays in 37 medical institutions in Japan using a Fricke dosimeter.

Materials and Methods

Table 1 summarizes the materials, methods and exposure factors employed in this study.

Dosimeter solutions were prepared by dissolving commercially available analytical and guaranteed grade chemicals without purification in triply distilled water.

The color caused by the reaction of ferric ions and thiocyanate ions was measured to determine the quantity of ferric ions produced by irradiation.

Fifteen dosimeters in glass tubes were mailed to 37 institutions; 10 for irradiation and 5 to act as non-irradiated controls. Two dosimeters placed in a lucite block phantom were used for each exposure. Chemical changes were assessed when they were returned. To check the sensitivity of dosimeters and to assess the chemical determinations, the dosimeter solutions taken from the same bottle sent to the institutions for irradiation were exposed to Co-60 γ-rays and the chemical changes were measured simultaneously.

G-values employed were 15.6 for 15 MeV electrons[6)] and 15.8 for 6 MV X-rays[7)] both of which are given in the literatures.

According to ICRU recommendation[8)], "R" should not be used as unit of radiation dose. However, at practically all institutions, doses were expressed in "R" unit obtained by thimble ionization chambers. In the comparison, absorbed doses by the Fricke dosimeters were converted to "R" using 0.91 as coefficient for electrons[9)] and 0.95 for X-rays[10)].

Table 1. Materials, Methods and Exposure Factors Used in the Intercomparison

	Electrons	X-rays
Dosimeter	Fricke Dosimeter Fe $(NH_4)_2(SO_4)_2$: 10^{-3}M, NaCl: 10^{-3}M, H_2SO_4: 0.8 N	
Irradiation Vessel	Glass Tube, 1.6cmϕ × 3.2cm	
Fe* Determination	Color Change by KSCN	
Energy	15 MeV	6 MV
Build up	5 mm	10 mm
Phantom	10×10× 2.8cm	10×10× 3.3cm
Dose	1500 rad	
Field	10×10 cm	

Results

1. Purity of Chemicals.

The chemical yield and reproducibility with the use of guaranteed grade ferrous ammonium sulfate and sodium chloride were of the same level as those with the use of chemicals purified by recrystallization. It has been reported that the effect of impurities from sulfuric acid could not be eliminated by the addition of sodium chloride and that pre-irradiation of sulfuric acid was effective[11]. No such effect was observed in sulfuric acid (Wako Pure Chemicals Co.) of analytical grade used in this study. Water was first passed through ion exchange resin and distilled thrice with alkaline and acidic potassium permanganate, and potassium bisulfate. The reproducibility of the dosimeter was less than $\pm 0.5\%$ at irradiation of about 5000 rads and the decrease in chemical yield by addition of sodium chloride was less than 1%, indicating that the purity of the dosimeter solution was good.

2. Thiocyanate Method for Fe^{+++} Determination.

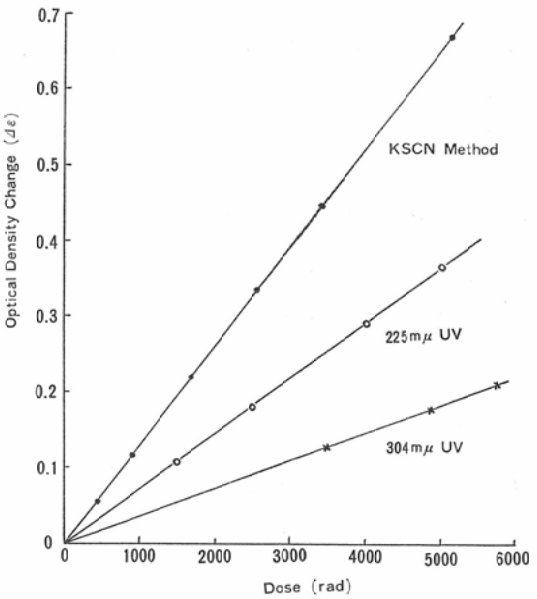
Doses of about 5000 rads are necessary to measure ferric ions by ultraviolet absorption at 305 mμ. Considering electron output from betatron, repeated exposures of such doses would pose some difficulty to busy institutions. In this study, thiocyanate method was employed to detect lower doses.

The relationship between dose and optical density by the potassium thiocyanate method is shown in Figure 1. Four milliliters of the dosimeter solution was mixed with 2 ml of 300 mg/ml potassium thiocyanate solution. Absorption at 472 mμ was measured 5 to 10 minutes after mixing.

The linearity obtained was satisfactory and accuracy was within 1.5% and slightly poorer than that of ultraviolet absorption at 304 mμ.

Lower doses can be detected by using higher concentration of potassium thiocyanate solution. According to Frigerio[12], it is possible to measure doses of 50 rads using solid ammonium thiocyanate, but we have found spontaneous oxidation of ferrous ions to occur at high concentrations. This has also been

Fig. 1 Dose-Optical Density Curve by KSCN Method
Co-60 γ-rays



pointed out by Fricke et al[13].

Figure 2 shows the thermal oxidation of ferrous ions by time after reaction. No change in optical density by time was observed up to 300 mg/ml solution, but optical density increased in highly concentrated solutions in both irradiated and non-irradiated dosimeters. At high concentrations, net changes in optical density were almost constant but fluctuations by time were larger. Two or three hundred rads can be measured by the use of saturated solution with an accuracy of about 5%, a level unsatisfactory for dose determinations.

Fig. 2 Optical Density by Time after Reaction

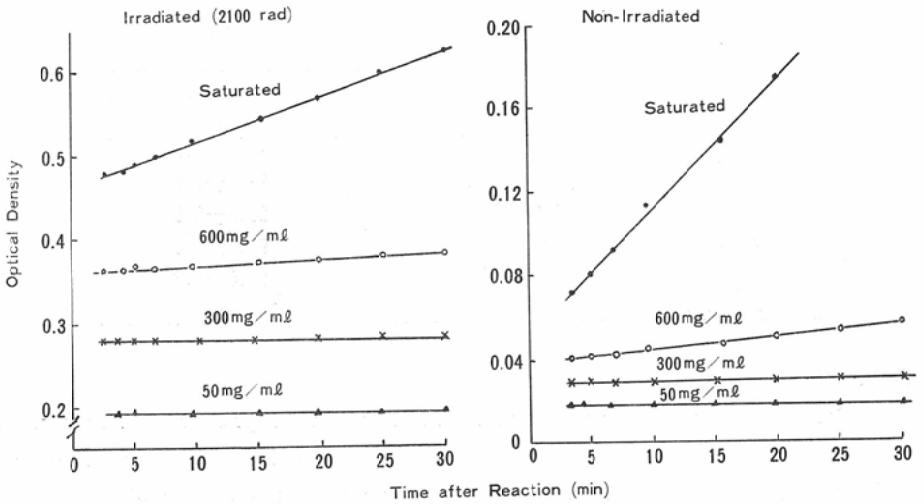
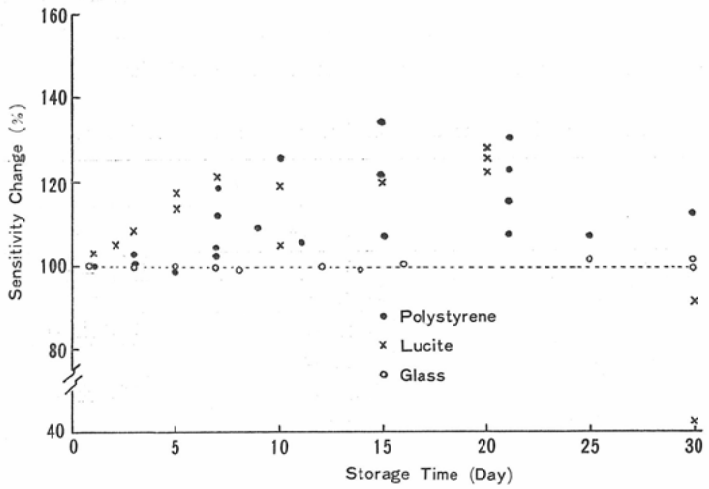


Fig. 3 Sensitivity Change of Dosimeter by Storage



3. Irradiation Vessel.

The irradiation vessel should be made of tissue equivalent material, such as polystyrene[14]. Weiss et al. have observed an elevated chemical yield using cylindrical glass vessels having internal diameters

of less than 8 mm for Co-60 γ-rays because of more secondary electrons from the glass wall than from the polystyrene wall[15]. However, the storage of dosimeter solution in plastic vessels brought about changes in sensitivity of the dosimeter as shown in Figure 3. No consistent increase was observed in plastic vessels and this effect could not be eliminated by pre-irradiating the vessel or by coating the vessel wall with silicon resin or wax. Contrary to plastic, no significant change in chemical yield stored in glass vessels was observed before and after irradiation.

Table 2 gives the percent ratio of chemical yield by a given dose in glass vessels in comparison with polystyrene vessels by radiation and energy. With high energy radiations, no significant variation in yield was observed by vessel materials. Therefore, pre-irradiated glass vessels were used in this study.

Table 2. Effect of Vessel Material on Chemical Yield

Radiation	Energy	Percent Ratio in Yield Glass/Polystyrene
X-rays	180 kV	98.9±0.1
γ-rays	1.17, 1.33MeV	99.8±1.2
Electrons	10 MeV	98.8±0.8
	15 MeV	99.5±1.4
	21 MeV	98.8±0.8
Neutrons	14 MeV	101 ±0.6

4. Effect of Sodium Chloride.

Sodium chloride is added to the dosimeter solution in order to suppress the enhanced oxidation of ferrous ions due to organic impurities. It has been reported that sodium chloride should not be added when assessing high dose rate[16]. Using sodium chloride, the yield decreased by about 1% for 10^{-3} M with Co-60 γ-rays. For electrons, the decrease was almost identical to that for Co-60 γ-rays as shown in Table 3. The average dose rate of electrons was about 500 rads/min while the true dose rate for pulsed radiation was about 1×10^5 rads/min. As the dose rate commonly used in electron therapy is less than 500 rads/min, 10^{-3} M of sodium chloride was added to dosimeter solutions.

Table. 3 Effect of Sodium Chloride on Chemical Yield

NaCl (M/l)	Percent Ratio in Yield	
	Co-60 γ-rays	Electrons
0	100 ±0.5	100 ±1.6
0.5×10^{-3}	99.0±0.4	—
1.0×10^{-3}	98.7±0.3	99.2±0.7
5.0×10^{-3}	98.6±0.2	—
1.0×10^{-2}	96.4±0.4	92.6±0.6
1.0×10^{-1}	92.8±0.2	—

5. Intercomparison of Electrons.

Table 4 shows the results of intercomparisons among 25 institutions participating in this study; 24 with betatron and 1 with linear accelerator.

The dosimeters used in these institutions are presented in the third column. Thimble chambers

were employed in all hospitals except for one institution where the type was unknown. The fourth column shows the institution dose to chemical dosimeters and the fifth, the average doses determined by the chemical dosimeters with standard deviations. Since the accuracy of the chemical dosimeters was within 1.5%, a standard deviation of less than 2% indicated good stablity of the dose monitor by time.

The sixth column shows the dose in "R" units converted from dose in rad and the seventh, the ratios of doses as determined by the institutions to the doses obtained by chemical dosimetry. The mean value of 25 institutions was 1.02.

Dosimetry conditions varied by institutions. In some institutions, ionization chambers were placed in phantom and in other institutions, they were placed in air. Chamber readings in air were 3.0% lower than those in phantom used. Doses provided by institutions were corrected for scattered radiation if dosimetry was carried out in the air. The last column gives the ratios of the corrected doses to the chemical doses. They averaged 1.01 ± 0.10.

Table 4. Intercomparison of Electrons from Betatron in Japan

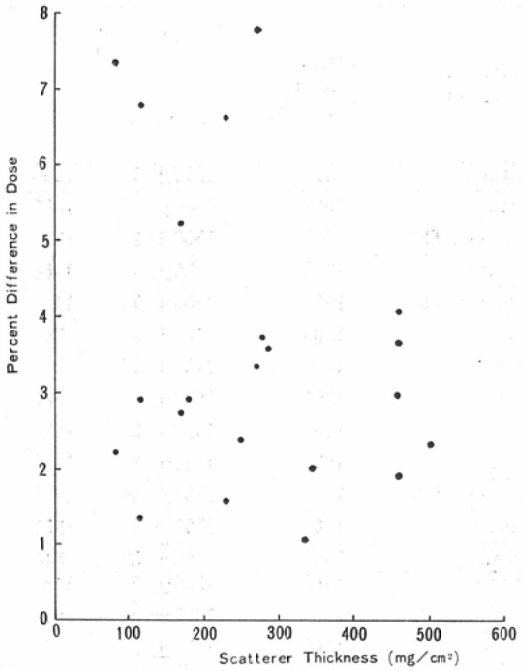
Institution	Energy (MeV)	Institution Dosimeter	Institution Chamber Dose (R)	Dose by Chemical Dosimeter		Chemical Dose / Chamber Dose in "R" Unit	Chemical Dose / Chamber Dose Corrected for Scattered Dose
				(M±σ) rad	(R)		
1	15	Radocon	1415	1270± 19	1390	0.96	0.93
2	15	Radocon	1555	1440± 9	1580	1.02	1.02
3	16	Victoreen 621	1500	1390± 29	1530	1.02	0.99
4	16	Radocon	1500	1330± 17	1460	0.97	0.95
5	16	Victoreen 621	1500	1350± 71	1480	0.99	0.99
6	15	Siemens	1500	1400± 26	1540	1.03	1.01
7	15	Home Made	1400rad	1400± 15	—	1.00	1.00
8	15	Ionex	1865	1620± 24	1780	0.95	0.95
9	15	Ionex	1500	1410± 26	1550	1.03	1.03
10*	10	Radocon	1622	1560± 52	1680	1.04	1.04
11	16	Victoreen 621	1500	1360± 44	1490	0.99	0.99
12	15	Radocon	1500	1390± 17	1530	1.02	1.02
13	15	Siemens	1500	1400± 37	1540	1.03	1.03
14	15	Radocon	1500	1350± 37	1480	0.99	0.99
15	16	Radocon	1500	1490± 31	1630	1.09	1.06
16	16	Radocon	1500	1530± 22	1680	1.16	1.16
17	15	Unknown	1500 ?	1000± 79	1100	0.73 ?	0.70 ?
18	16	Ionex	1500	1200± 17	1320	0.88	0.88
19	15.4	Victoreen 621	1500	1270± 33	1400	0.93	0.93
20	16	Siemens	1500	1560± 41	1710	1.14	1.11
21	16	Radocon	1617	1450± 30	1590	0.98	0.98
22	15	Siemens	1500	1340± 17	1470	0.98	0.95
23	14	Siemens	1500	1690± 81	1860	1.24	1.21
24	16	Victoreen 621	1500	1430± 23	1580	1.05	1.05
25	16	Radocon	1500	1630±110	1790	1.19	1.19
					Mean	1.02	1.01±0.10

* Linear Accelerator, $C_2=0.93$

Dose determinations should be as accurate as possible. Acceptable limit of accuracy should perhaps be within 10% in radiation therapy. As shown in the table, six of the 25 institutions exceeded the 10% limit of accuracy, 80% of the institutions being within the limit. Relative dose values were in good agreement among the institutions despite the theoretical problems existing in high energy electron dosimetry.

Two dosimeters in each exposure were placed 1.3 cm to the right and left of the beam center of the 10 × 10 cm field. Relationship between dose homogeneity and scatterer thickness is shown in Figure 4. The vertical scale shows the percent differences in average chemcal yields between the right and left dosimeters irradiated simultaneously and the horizontal scale shows the thickness of scatterer in mg/cm^2. Correlation factor showed statistically insignificant value of 0.25. If unhomogeneity is symmetrical, difference can not be detected by this analysis. However the thinner the scatterer in general the larger became the differences. A detailed study of field homogeneity by scatterer has been reported by Rassow[17].

Fig. 4 Dose Homogeneity in Field by Scatterer Thickness



There were no particular correlations between the accuracy of chamber readings and the manufacturer, indicating that changes in sensitivity may be caused through the usage and storage of the chamber.

6. Intercomparison of X-rays.

Intercomparison of X-ray doses from 12 linear accelerators is shown in Table 5. The columns are similar to those in the table 4.

The average ratio in 12 institutions was 1.01 ± 0.09 and 3 of the 12 institutions exceeded the 10% limit of error, similar to the case of electron dose determinations.

Table 5. Intercomparison of X-rays from Linear Accelerator in Japan

Institution	Energy (MV)	Institution Dosimeter	Institution Chamber Dose (R)	Dose by Chemical Dosimeter		Chemical Dose / Chamber Dose in "R" Unit
				(M±σ) rad	(R)	
1	6	Ionex	1500	1417±14	1530	1.02
2	5.7	Victoreen 621	1500	1330±21	1380	0.92
3	8	Radocon	1500	1270±43	1330	0.88
4	10	Radocon	1514	1420±16	1480	0.98
5	6	Siemens	1500	1680±16	1750	1.16
6	6	Ionex	1500	1420±37	1480	0.99
7	6	Ionex	1640	1560±13	1620	0.99
8	6	Unknown	1500?	1660±37	1730	1.15?
9	6	Radocon	1500	1410±21	1470	0.98
10	6	Victoreen 621	1513	1590±21	1650	1.10
11	6	Siemens	1500	1480± 8	1540	1.03
12	6	Radocon	1500	1350±35	1410	0.94
					Mean	1.01±0.09

Discussion

The use of Fricke dosimeter for intercomparisons is complicated by the sensitivity changes through storage of the dosimeter in plastic vessels and by the necessity of using large dose to attain accuracy.

Petterson et al.[18] have also observed the sensitivity changes in plastic vessels. Pre-irradiation of vessels has been found to be ineffective in eliminating of storage effect. Pre-irradiation may supply the organic degradated substances from polymer in the case of polystyrene and lucite, the representative polymer which degrade by irradiation. Although coating of wall by silicon resin, having cross-linking nature by irradiation, seemed to be effective, the softening of plastic materials at relatively low temperature precluded coating by heat. Glass tubes were used because no sensitivity change could be observed through storage and because variation in yield between glass and polystyrene was insignificant.

IAEA has reported that the standard deviation for dose determination by a single Fricke ampoule was about 0.2%, compared to about 1.4 for a single LiF capsule[3]. In our experience, a dose of about 5000 rads was necessary to obtain a standard deviation of less than 0.5%. The repeated exposure of 5000 rads may be pose an excessive burden for some institutions. The accuracy of the Fricke dosimeter used in this study was slightly poorer than that of the standard dosimeter because a modified Fricke dosimeter was used in order to detect lower doses. As the sacrifice of accuracy in the dosimeter itself, comparisons were repeated in five sets of dosimeters. This procedure reduced the error involved in setting the dosimeters in exposure field and the error in chemical determinations and permitted checking of the stability of the dose monitors attached to generators.

G-values and conversion factors from "R" to rad are not important for intercomparison purposes. Aside from the question of intercomparison, it is possible to obtain doses in absolute unit with Fricke dosimeter by means of G-values given in the literature. Shalek et al.[19] have reported the G-values to be 15.5 for electrons and 15.7 for X-rays. These values were slightly lower than those employed in this study. However, the difference is less than 1%, a value which poses no practical problem.

Some data are available concerning the conversion factors[20][21][22]. Matsuzawa et al. have calculated

the factor to be 0.91 for 15 MeV electrons[9] based on energy spectra in water phantom. ICRU Table[10] gives a value of 0.95 for X-rays. The study of conversion factors is in progress and it may be possible that these values will be changed within a few percent.

The sensitivity changes of ionization chambers by radiations and their energies are still within the scope of conjecture. It has been reported that the magnitude of such changes is different by electrons and X-rays, and by chambers.[28] If this is so, chambers calibrated for Co-60 γ-rays can not be used for electrons and X-rays without correction and the discrepancy of doses among institutions may be ascribable to such sensitivity changes. Although no change was observed in our Radocon, probe 607, by electron energy as shown in Table 6, more reliable data should be accummulated on this subject.

Table 6. Sensitivity Change of Ionization Chamber by Radiation and Energy

Radiation	Energy (MeV)	Percent Ratio $\left(\frac{\text{Radocon Dose}}{\text{Chemical Dose}}\right)$		Mean
		I	II	
Electrons	21	99	97	98
	15	100	100	100
	10	98	98	98
Co-60 γ-rays	1.17, 1.33	100	100	100

This study included the majority of therapeutic units in Japan, namely about 35 betatrons and 20 linear accelerators being in use late 1968. Although the institutions participating in this study differed by electrons and X-rays, the average comparison ratio and standard deviation were similar. This suggests that the accuracy of dosimetry in other institutions may be of the same order as that in this study. Many dosimeters in medical institutions have been calibrated by Hashizume et al. for Co-60 γ-rays. This may be one of the reasons for good agreement among institutions.

In comparison with ionization chambers, the Fricke dosimeter has many advantages, including low energy and dose rate dependency and tissue equivalence. However, from the practical standpoint, large doses are necessary and some inconveniences are involved in preparations of solution and chemical determinations. Although chemical dosimeters are useful in dose comparisons, their best use in the calibration of ionization chambers and in the verification of dose measured by other apparatus, such as thermoluminescence and fluoroglass dosimeters.

Summary

Intercomparison of high energy electrons and X-rays in 37 medical institutions in Japan was made by using a Fricke dosimeter.

The color developed by the reaction of ferric ions and thiocyanate ions was measured to determine the quantity of ferric ions. This method permitted determination of doses lower than by ultraviolet absorption at 304 mμ and 225 mμ. The accuracy of the method was less than 1.5% and slightly poorer than that by ultraviolet absorption.

Sensitivity of dosimeter increased by the storage in polystylrene and lucite vessels. Glass tubes were

used as irradiation vessels because storage did not introduce any sensitivity change and because chemical yield was similar to plastic vessels.

The average ratio of doses in participating institutions to the doses evaluated by Fricke dosimeters was 1.01 ± 0.10 for electrons and 1.01 ± 0.09 for X-rays. Good agreement in dose among the institutions was obtained for both electrons and X-rays.

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Reference

1) Hashizume, T. and Maruyama, T.: Ann. Rep. Minist. Educ. 1965, Radiation Effects-Coop. Res. p. 177, 1966.
2) Kato, Y., Shiragai, A., Kumamoto, Y., Maruyama, T., Yoshida, Y. and Hashizume, T.: Nipp. Act. Radiol., 29, 7, 1969.
3) Loevinger, R., Pfalzner, P.H., Eisenlohr, H. and Malo, S.A.: Annals of the New York Academy of Sciences, 161, Art 1, 158, 1968.
4) Pfalzner, P.M. and Jayarama, S.: Presented at XIIth ICR, Oct. 6—11, 1969, Tokyo, Japan. Book of Abstract, XIIth ICR. p. 454, 1969.
5) Laughlin, J.S., Shalek, R.J., Ovadia, J. and Holt, J.G.: Phys. Med. Biol., 10, 429, 1965.
6) Petterson, C.: Arkv For Fysik, 34, 385, 1967.
7) Shalek, R.J. and Smith, C.E.: Annals of the New York Academy of Sciences, 161, Art 1, 44, 1965.
8) NBS Handbook 84, Radiation Qualities and Units, ICRU Report 10a, p. 2, 1962.
9) Matsuzawa, H., Inada, T. and Hoshino, K.: Ann. Rep. Minist. Educ., 1968, Radiation Effects, Academic Press, p. 32, 1969.
10) NBS Handbook 85, Physical Aspects of Irradiation, Recommendation of ICRU, Report 10b, p. 3, 1962.
11) Davis, J.V. and Law, J.: Phys. Med. Biol., 8, 91, 1963.
12) Frigerio, N.A. and Henry, V.D.: Solid State and Chemical Radiation Dosimetry in Medicine and Biology, IAEA, 257, 1967.
13) Fricke, H. and Hart, E.J.: Radiation Dosimetry II, edited by F.H. Attix and W.C. Roesch, Academic Press p. 197, 1966.
14) Loevinger, R., Karzmark, C.J. and Weissbluth, M.: Radiology, 77, 906, 1961.
15) Weiss, J., Allen, A.O. and Schwarz, N.A.: Proc. 1st Intern. Conf. Peaceful Uses of Atomic Energy, Geneva, UN, 14, 179, 1955.
16) Thomas, J.K. and Hart, E.J.: Radiat. Res., 17, 408, 1962.
17) Rassow, J.: Acta Radiol. Ther. Phys. Biol., 8, 331, 1969.
18) Petterson, C. and Hettinger, G.: Acta Radiol. Ther. Phys. Biol., 6, 160, 1967.
19) Shalek, R.J., Sinclear, W.K. and Calkins, J.C.: Radiat. Res., 16, 344, 1962.
20) The Hospital Physicist's Association: Phys. Med. Biol., 14, 1, 1969.
21) Almond, P.R.: Phys. Med. Biol., 12, 13, 1967.
22) MacDonald, J.C.F., Kartha, M., Dawson, D.J. and Oldfield, L.C.: Presented at XIIth ICR, Oct. 6—11, 1969, Tokyo, Japan, Book of Abstract, XIIth ICR, p. 454, 1969.
23) Hiraoka, T., Habu, T. and Matsuzawa, H.: Nipp. Act. Radiol., 27, 1315, 1968.