MEASUREMENTS OF ^{32}P IN SULFUR

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The type of Lauritsen electroscope used by Yamasaki and Sugimoto in their sulfur measurements is not yet finally identified. At the first Dosimetry Workshop in Nagasaki a picture of an electroscope was shown, with a chamber volume of 368 cm, which was found in the laboratory and thought to be the one commonly used at that time. However, it was apparently hand made with an inlet and outlet for gas, and seems to have been used for a particular experiment.

Afterwards, an electroscope of another type was discovered in the laboratory (Figure 1) which appears to be the same type as one seen in a film "Effects of the Atomic Bomb." It is made of cast aluminum and seems to have been manufactured as a commercial product. It is known that Dr. Yamasaki had tested seven electroscopes and used the one with the highest sensitivity.[1] This suggests that the latter type was the one used by Yamasaki and Sugimoto. The chamber volume of this electroscope is 111 cm^3.

The Lauritsen electroscope is designed to have the highest sensitivity, at the expense of stability, for use mainly in activity measurements, unlike the conventional electroscope for dosimetric use such as with the Victoreen r-meter. Its sensing element consists of a quartz fiber 3μm in diameter and 10 mm long supported at one end, and its indication is very sensitive to vibration and air convection in the chamber. The element has an electrical capacity of only 0.3 pF.

Recently more repeated measurements of a KCl sample were carried out which give readings of about twice the background at room temperature ranging from 25 to 27°C. The standard deviation of readings was almost the same as reported at the second Dosimetry Workshop in Hiroshima.[2]

It was seen that, in most measurements, the first readings were significantly larger than the following ones. This suggests absorption of the charge into the insulator had occurred (i.e., polarization of the insulating material). This effect, rather than fluctuations of ionization

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This article is a composite of a note sent to the US Committees for Reassesement of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki in December 1984, and a letter sent to W.C. Roesch on 17 June 1985.

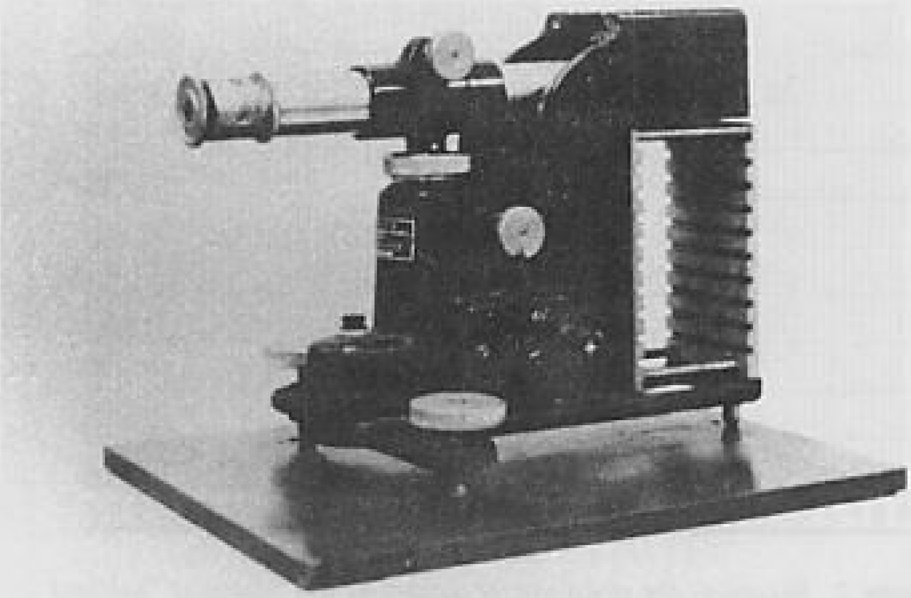

Figure 1. Lauritsen electroscope recently discovered in the laboratory

in the chamber, would probably be the main cause for large fluctuations of the electroscope readings.

Uncertainty of Electroscope Readings

The contribution of alpha particles to the electroscope background and the single-event charge distribution for alpha particles were investigated by Dr. Hamada on an electroscope considered to be the same type as that used by Yamasaki and Sugimoto for determining ^{32}P activity induced in sulfur. The results showed that the uncertainties of the electroscope readings agreed with those reported previously.[2]

The ion chamber of the electroscope used by Dr. Hamada has an inner dimension of $5.1 \times 5.3 \times 4.1$ cm and a volume of 111 cm^3. It is made of cast aluminum and the inner surface is coated with colloidal graphite except for the bottom window of aluminum foil. It does not work as an electroscope on account of lack of the quartz fiber. The ionization in the chamber was measured by inserting a collector electrode connected with a vibrating reed electrometer through a hole in the rear side.

Results and Discussion

Contribution of alpha particles to the electroscope background. According to the background data of Yamasaki and Sugimoto, their measurements seem to have been made without shielding the electroscope. Therefore, the background ionization was measured by the "charging" method without shielding.

A part of the recording chart is shown in Figure 2. The sharp rises correspond to the charge produced by individual alpha particles and are identified visually on the chart. The result of five runs of measurement by the "charging" method are given in Table 1. Ten divisions correspond to 60 mV and the input capacity of the electrometer is 10.54 pF, therefore, the ionization for one minute was:

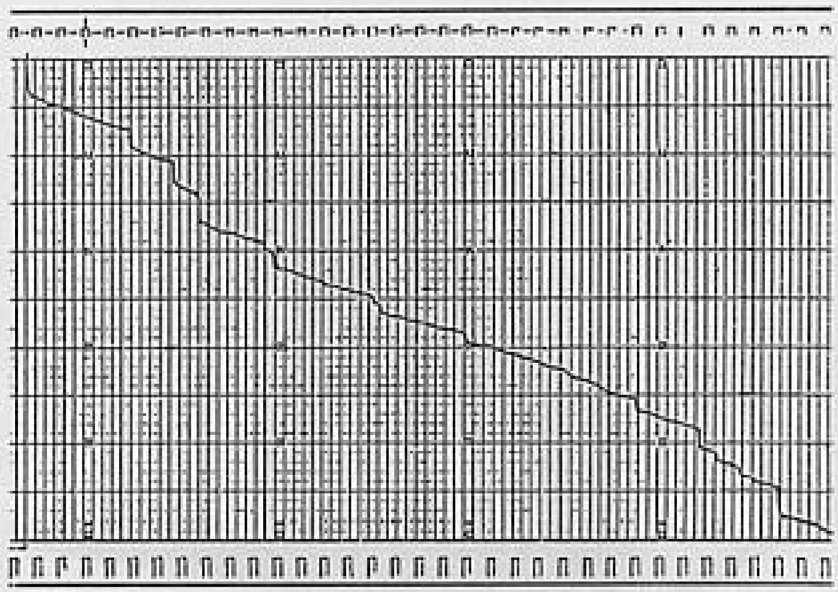

Figure 2. Record of the electrometer output by the "charging" method

Table 1. Results of Measurements by the "Charging" Method

Run	Time (min)	Ionization (div)	
		Total	Alpha
1	38	9.29	4.78
2	30	8.93	5.02
3	30	8.95	5.28
4	30	9.20	5.59
5	30	8.98	5.11
Total	158	45.35	25.78

Total ionization per min, (q_B): 1.12×10^5 ion pairs per minute,
Alpha ionization per min, $(q_B)_\alpha$: 6.39×10^4 ion pairs per minute.

The value of $(q_B)_\alpha$ would be somewhat underestimated because small ionizations produced by single alpha particles could not be identified on the record and were neglected.

Single-Event Change Distribution for Alpha Particles. Variation of ionization current of the electroscope was measured by the "current" method. From the height of alpha particle peaks on the recording chart and the value of $(q_B)_\alpha$ (above), the sum of squares of a single-event charge produced by alpha particles, $(q_B{}^2)_\alpha$ was calculated. A part of the record obtained by the "current" method is reproduced in Figure 3, where the height of each peak corresponds to a single-event charge produced by an alpha particle. A histogram of the pulse-height distribution is shown in Figure 4. The results of the measurement are: time of measurement, 488 min; number of alpha pulses, 540; sum of peak height of all alpha particle pulses, 884.11 div; and sum of squares of peak heights of alpha pulses, 2090.57 $(\text{div})^2$.

Since the sum of the peak height of alpha particle pulses should be equal to the product

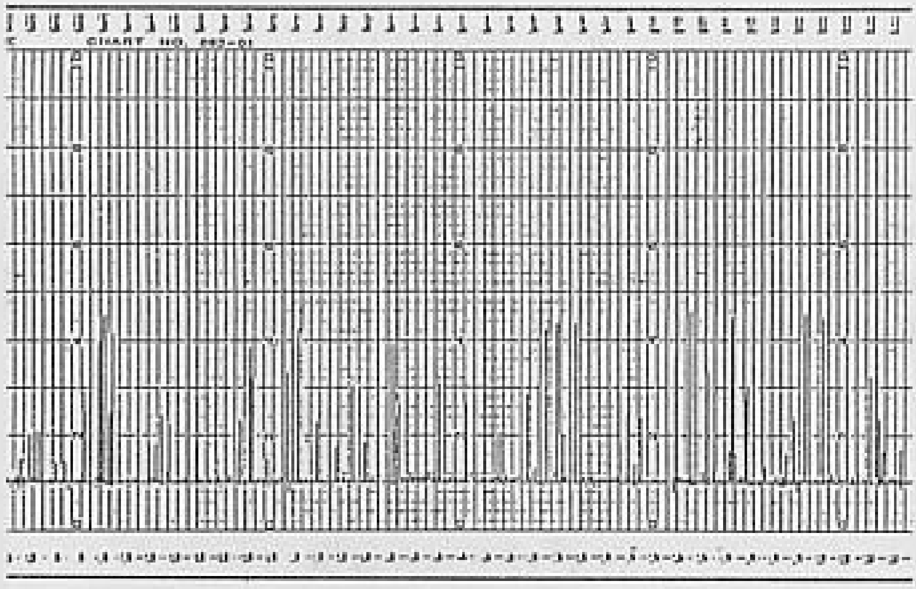

Figure 3. Record of the electrometer output by the "current" method

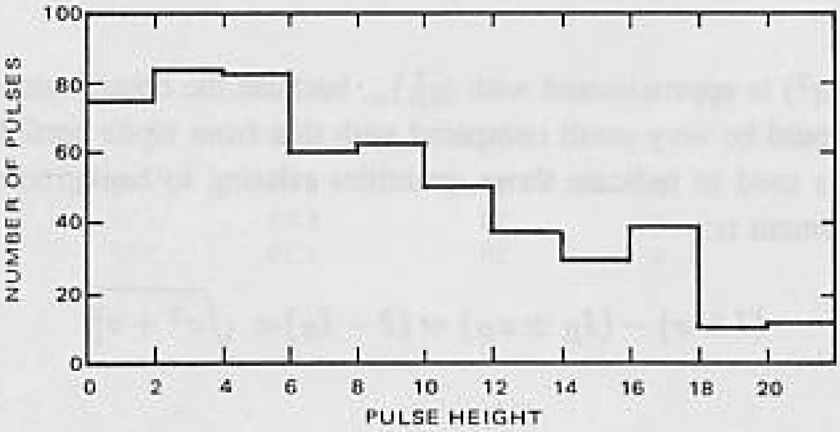

Figure 4. Histogram of the alpha-particle pulse heights

of the duration of the measurement and $(q_B)_\alpha$, then 884.11 div = 488 min $\times 6.39\times 10^4$ ion pairs per minute. Therefore, one division corresponds to 3.53×10^4 ion pairs. Thus, the sum of squares of the peak heights of alpha particles per minute, $(q_B{}^2)_\alpha$, is $(q_B{}^2)_\alpha = 5.34\times 10^9$ (ion pairs)^2 per minute.

Estimation of the Standard Deviation of Electroscope Readings. The method for determining the standard deviation of the fluctuations in the charge collected from an ionization chamber from the mean and mean-square of the charges deposited in individual events is known.[3] The method, however, is for application to charges collected in equal times. Yamasaki and Sugimoto used a method, common at the time, of measuring the times to collect equal charges. Roesch and Jablon independently showed that for the magnitudes of the charges dealt with in the Yamasaki-Sugimoto apparatus the result was the same as for the better known theory (personal communications). The standard deviation(σ), of the time (t), required for collection of a charge Q is:

$$\frac{\sigma}{t} = \sqrt{\frac{< q^2 : 1 >}{< q : 1 > Q}} \tag{1}$$

where <q:1> and <q^2:1> are the first and second moments of the single-event charge distribution. Since the elctroscope reading I in div/s is proportional to Q/t, the relative standard deviation of the reading σ/I is equal to σ/t.

If the particles enter the electroscope at a rate of r (min^{-1}), then

$$Q =< q : 1 > rt = (q)t \tag{2}$$

Similarly,

$$< q^2 : 1 > rt = (q^2)t \tag{3}$$

Then,

$$\frac{\sigma}{I} = \frac{\sigma}{t} = \sqrt{\frac{< q^2 : 1 >}{< q : 1 > Q}} \tag{4}$$

In Equation (4), (q^2) is approximated with $(q_B^2)_\alpha$, because the contribution of electrons and mesons to (q^2) would be very small compared with that from alpha particles.

Subscript B is used to indicate those quantities relating to background. Then, the net result of a measurment is:

$$(I \pm \sigma) - (I_B \pm \sigma_B) = (I - I_B) \pm \sqrt{\sigma^2 + \sigma_B^2} \tag{5}$$

In Equation (4),

$$I = \frac{Q}{t} = (q) \tag{6}$$

and

$$\sigma = (q)\sqrt{\frac{(q_B^2)_\alpha}{(q)^2 t}} = \sqrt{\frac{(q_B^2)_\alpha}{t}} \tag{7}$$

Therefore, the relative standard deviation of the net result is

$$\frac{\sqrt{\sigma^2 + \sigma_B^2}}{I - I_B} = \frac{\sqrt{(q_B^2)_\alpha}\,[\frac{1}{t} + \frac{1}{t_B}]^{1/2}}{(x - 1)\,(q_B)} \tag{8}$$

where

$$x = \frac{(q)}{(q_B)} = \frac{I}{I_B} \tag{9}$$

Table 2. Standard Deviations of Electroscope Readings

Sample	Time for Measurement (min)	Electrometer[a] Reading (div/s)	Relative Standard Deviation
Background	200	0.00124	0.06
A-4	40	0.00200	0.19
B-5	38	0.00224	0.14
C-6	45	0.00154	0.45
D-12	70	0.00162	0.29
D-12	70	0.00178	0.21
E-13	95	0.00205	0.13
F-14	74	0.00166	0.26
G-15	90	0.00167	0.24
G-15	90	0.00175	0.20
H-7	85	0.00144	0.53
J-8	103	0.00130	1.58
K-28	b		

a. From Yamasaki's record.
b. No data given.

The relative standard deviations of the sulfur ^{32}P activity measurements were calculated from Equation (8). The results are given in Table 2.

Sulfur Purity

Two new studies in addition to the one reported by Hamada[2] have been made of the purity of the sulfur used by Yamasaki and Sugimoto.

Proton-Induced X-ray Emission. H. Takeshita, S. Kawamura, K. Kitao, and T. Maruyama of the National Institute of Radiological Sciences analyzed some of the sulfur samples collected in Hiroshima by Yamasaki and Sugimoto for their sulfur ^{32}P activity measurements. Sulfur content of the samples was determined by volumetric analysis and their impurity elements by proton-induced x-ray emission (PIXE) analysis.

A known amount of the sample was dissolved in acetone, diluted with a 1/5 amount of water, heated to a temperature just below boiling, and titrated with a isopropyl alcohol solution of NaCN using bromocresol purple as an indicator. The results are shown in Table 3 together with the data previously obtained by Hamada.[2]

A small amount of pulverized sample was bombarded by 2.5 MeV protons from a Van de Graaff accelerator for 10 to 15 minutes and the x rays emitted were detected by a Si(Li) detector. The energy spectra show the presence of several heavy elements as listed in Table 4. However, these impurities, activated with neutrons, do not seem to interfere significantly with the sulfur ^{32}P activity measurements.

Neutron Activation Analysis. About 50 mg of CS_2-insoluble fraction of sulfur samples were activated in a reactor with a thermal neutron fluence of 6×10^{16} cm^{-2}. Ten days after the neutron irradiation, the activated product was subject to gamma-ray spectroscopy. The main radionuclides detected and their activity are shown in Table 5.

The thermal neutron fluence at the hypocenter in Hiroshima is estimated at about 10^{12}

Table 3. Results of Volumetric Analysis of Sulfur Samples

Sample[a]	Sulfur Found (%)				Remaining Fraction (%)	
	Run 1	Run 2	Run 3	Average	This Work	Hamada[2]
1	100	100	100	100	0	0
2	98.1	99.3	99.2	98.9	1.1	1.61
3	98.1	98.1	99.7	98.6	1.4	
J-8						0.27
11	80.0	80.6	81.4	80.7	19.3	
15	99.0	99.0	100	99.3	0.2	
16						2.62
19						1.67
26	100	100	100	100	0	2.07
K-29	55.3	55.6	54.1	55.0	45.0	35.3

[a] Numbered by Yamasaki.

Table 4. Results of Proton-induced X-ray Emission Analysis of Sulfur Samples

Element	Sample No.						
	1	2	3	11	15	26	K-29
S[a]	100 %	98.9%	98.6%	80.7%	99.3%	100 %	55.0%
K	++++	++++	++++	D	++++	++++	0.5%
Ca	++++	++++	++++	7 %	++++	++++	10.0%
Ti	+++	?	D	–	++++	D	+++
Cr	++	D	–	–	–	++	D
Mn	?	D	–	D	++	++	+++
Fe	++	+++	+++	++++	++++	++++	0.2%
Ni	+	–	–	–	–	–	–
Cu	++	D	–	–	+	++	+
Zn	++++	++++	++++	++	++++	+++	++++
As	+	++	++	D	D	+	+
Se	++	+++	++	++	++	++	++
Rb	D	–	–	–	–	–	–
Sr	D	–	–	++	–	–	++
La	?	D	D	–	–	D	–
Pb	D	D	–	–	–	D	–

[a] From volumetric analysis.

++++	0.05 - 0.01%	+	0.0009 - 0.0004%
+++	0.0009 - 0.005%	D	detected but could not determine
++	0.004 - 0.001%	–	not detected

cm^{-2}. The ^{32}P activities in 1 g of sulfur samples determined by Yamasaki and Sugimoto 35 to 37 days after the explosion were larger than 1 Bq except for J-8.[2] Therefore, on the assumption that the sulfur samples contained impurities as much as 5% by weight, the interference of these activated impurities with the ^{32}P activity measurements would be negligible.

Rejected Data for ^{32}P in Sulfur. Hamada[1] describes how Yamasaki and Sugimoto measured 13 sulfur samples, 3 of which (samples 1, 3, and 16) were rejected because of lack of

Table 5. Results of Neutron Activation Analysis of Sulfur Samples

Radionuclide	Half Life	Activity (Bq/50 mg) after	
		10 days	35 days
^{46}Sc	83.8 d	73	60
^{47}Ca + ^{47}Sc	4.54 d	83	1.8
^{59}Fe	44.6 d	36	24
^{65}Zn	244.1 d	1640	1530
^{76}As	26.3 h	2560	3.5×10^{-4}
^{75}Se	118.5 d	6.7	5.8
^{82}Br	35.4 d	10	8×10^{-5}
^{124}Sb	60.2 d	27	20

Table 6. Data on Rejected Observations of ^{32}P in Sulfur

Sample No.	Time of Measurement	Electroscope Reading (div/s)		^{32}P Activity (Bq/g S)	Location	Ground[c] Distance (m)
		Gross	Net[a]			
1	10/IX 14th 03 min	0.0013	0.00006	0.53±1.0[b]	45.67 × 61.35	1305
3	10/IX 14th 58 min	0.0013	0.00006	0.53±1.0	45.05 × 61.60	705
16	12/IX 15th 50 min	0.0012	0	0 ±1.0	44.23 × 60.64	967

[a] Background: 0.00124 div/s.

[b] Estimated standard deviation at the time of measurement.[2]

[c] Based on the new RERF (T65D) hypocenter coordinate (44.285 × 61.697).

significant activity. Table 6 gives the data for these samples. Using the efficiency value[2] of 1.14×10^{-4} divisions per disintegration gives the data for the rejected samples shown in Table 6. These should be added to Table 2 in Hamada's paper.[2]

Location and ground distance for these 3 rejected samples were determined by RERF based on Yamasaki's original map and on the new RERF hypocenter coordinates used in the T65D dose estimation. The results are given in Table 6.

References

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2. Hamada, T., ^{32}P activity induced in sulfur in Hiroshima: reevaluation of data by Yamasaki and Sugimoto. In *Second U.S.-Japan Joint Workshop for Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki*,pp. 52-55. Hiroshima: Radiation Effects Research Foundation.
3. International Commission on Radiological Units and Measurments, 1983. *Microdosimetry*, pp. 70-71. Bethesda, MD: ICRU report 36.