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On the Nature and Composition of Long-range Fallout

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SINGLE PARTICLE OF HIGH ACTIVITY

For over two years (since May 1956) a daily survey of fallout contamination, using the air filter method, has been carried out in Rio de Janeiro. The filters (TFA # 2133) are rectangular in shape having an area of 51 cm²; 400 to 600 m³ of air are passed through them in a 12 hr sampling period; retention efficiency for 0.3 μ particles at 250 cm/sec flow-velocity is 99%. The activity of the filters is measured 4 to 6 days after sampling by wrapping them around cylindrical β counters with Al wall 0.15 mm (40 mg/cm²) thick.

The filter of 25 June 1957 showed an unusual strong activity, about ten times higher than the most active filters commonly observed. The first measurement,[1] 6 days after sampling, gave 1211 counts/min. Measuring the decay curve for several months (see Fig. 1) and applying Way and Wigner's law,[2] the origin of this activity could be traced back to the British H-bomb test in the Christmas Islands on 15 May.[1] The decay curve for fission products can be represented by the expression:

$$I(t) = c(t + t_0)^{-n},\ n \simeq 1.2 \qquad (1)$$

In this equation $(t + t_0)$ is the time between explosion and measurement; thus t_0 is the time between explosion and sampling. In a double logarithmic scale the decay curve must be a straight line, provided the right value for t_0 is chosen.

In the present case, applying a trial and error method, $t_0 = 50$ days is found to give a straight line (see Fig. 1). Also, for n the value 1.3 results. The sampling date was 25 June 1958; subtraction of 50 days leads to 6 May as the approximate date of explosion. According to newspaper reports there was an H-bomb explosion on 15 May in the Christmas Islands; this was the first British H-bomb test. Both dates are sufficiently close together to suggest that the collected activity was actually due to the British test explosion. This is confirmed also by the fact that the sampling date of the filter coincides with the front wave of a general rise in fallout activity, lasting several weeks.

The uniqueness of such high a value (high at least for southern latitudes) suggested the possibility of attributing almost all of its activity to a single particle of more than usual size. This plausible assumption could be confirmed by direct measurements.

On dissecting the filter into 4 strips and measuring each section individually, the following counts were obtained: 1.5, 1.6, 5.5 and 268 counts/min. Therefore, practically, the entire activity was concentrated on the last section of the filter. The existence of a single particle was brought out definitely by taking an autoradiograph, leaving the filter for 5 days in direct contact with a photographic plate (Gevaert, Gevechrom 32). (See Fig. 2.)

If the efficiency of the measuring equipment is determined (as $\simeq$10%) by means of a standard β source of appropriate energy and the self-absorption of filter taken into account ($\simeq$21%; see Appendix I) the absolute activity of the filter can be calculated. An approximate value of 5×10^{-9} curie for the day of the first measurement was found.

In addition, an approximate value for the size of the particle can be obtained as follows: if $N(t)$ is the total number of radioactive atoms and $(dN/dt)_0$ the activity at $t = 0$, i.e., at the day of the first measurement, then Way and Wigner's law is equivalent to

$$I(t) = \frac{dN(t)}{dt} = \left.\frac{dN}{dt}\right|_0 \times \frac{t_0^n}{(t + t_0)} \qquad (2)$$

Integrating between 0 and ∞ gives for $N(0)$ the value

$$N(0) = \left.\frac{dN}{dt}\right|_0 \times \frac{t_0}{(n - 1)}. \qquad (3)$$

Observing that here $(dN/dt)_0$ is the value of measured activity corrected for equipment efficiency and self-absorption, one obtains, by substituting the measured values, $N(0) = 1.6 \times 10^9$ atoms. Assuming for the fission-products a medium atomic weight of 144, corresponding to cerium, the weight of the particle would be approximately 4×10^{-13} g. The specific mass of cerium is 6.8 g/cm³; therefore, the volume of such a particle is approximately 6×10^{-14} cm³ and its radius equal to 1/4 μ on the day of the first measurement, nearly two months after the explosion. This estimate, of course, refers solely to the "active" mass of the particle.

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Although Way and Wigner's theoretical decay curve follows quite closely the experimental one of Hunter and Ballou,[3] the size determined is clearly a lower limit only, since most likely, not all of the decay products are present. Besides, the assumption of a particle of exclusively "active" mass is an over simplification, not taking into account condensation processes of fission products on some aerosol nucleus at the time of the explosion or afterwards.

To investigate further both the nature and activity of this particle, an absorption curve was determined, a comparison test was performed and an absolute calibration of its size and activity was carried out.

ABSORPTION MEASUREMENT

Using an end-window Geiger counter of 1.4 mg/cm² and a set of calibrated Al absorbers in always identical geometrical arrangements (source and filter as close to the window as possible), an absorption curve under good statistical conditions was obtained (relative statistical error $<1\%$ and $<10\%$ at the upper and lower end of the curve respectively).

The appearance is that of a typical β absorption curve with several different energies present (Fig. 3). The actual form of such curves depends on the geometry and sensitivity of the counter arrangements and of the source. The measured curve represents the superposition of a divergent beam of rays and easily conceals the existence of groups of rays with different energies.

For a point-source situated along the axis of symmetry at a fixed distance from a cylindrical end-window Geiger counter the application of the Gross

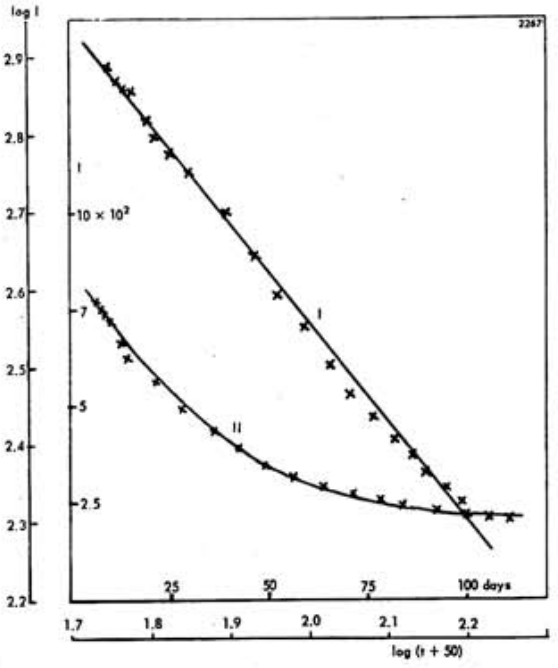

Figure 1. Decay curves. I—Log of number of counts/min against log $(t + 50)$. II—Number of counts/min against time in days

Figure 2. Autoradiograph of filter section containing radioactive fragment (exposure 5 days, natural size)

transform[4] is possible. In this way, the absorption curve for a parallel beam is obtained.

The original application of the transform in the theory of cosmic rays corresponded to a problem with 2π geometry. In this case a solution in closed form can be given. In the present case the solid angle between source and counter is less than 2π. A difference equation results which is not solved in closed form. A discussion of the equation has been given by Gross (UNESCO Symposium on Cosmic Rays, Cracow, 1947, unpublished). An iteration method can be applied (see Appendix II), which gives for the absorption curve of a parallel beam the expression

$$\psi(x) = \sum_{0}^{\infty} a_n f(x/a^n) \tag{4a}$$

$$f(x) = \frac{1}{2\pi}\left[J(x) - x\frac{dJ(x)}{dx}\right] \tag{4b}$$

where $a = \cos\theta$, and 2θ the angle of aperture. $J(x)$ is the measured absorption curve. For the geometry of the measuring arrangement employed in our measurements $\theta_0 = 59.6°$ and thus $a = 0.506$. The expression converges rapidly. In the present case 3 terms are sufficient. The transformed curve is shown in Fig. 4.

On this transformed curve, a separation into several different energy components is clearly apparent. An almost complete lack of γ radiation is a further distinctive feature of this curve. Even so, any precise energy determination by use of Feather's rule[5] or of

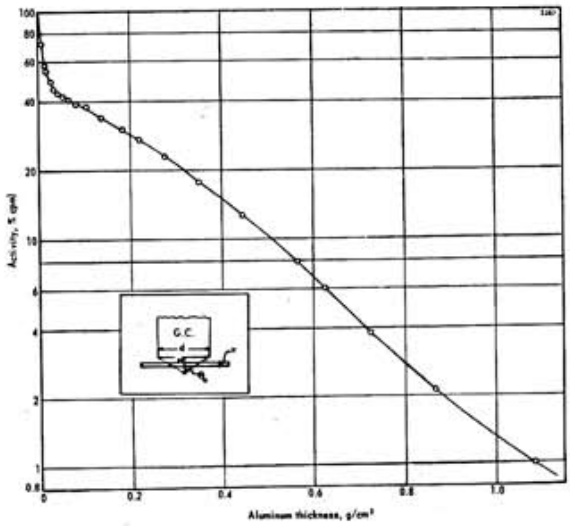

Figure 3. Al absorption curve of radioactive filter-particle. Inset: Geometry of absorber arrangement (H = 8.5 mm; $\theta_0 = 59.6°$); (x = Al − absorber thickness)

Glendenin's curves[6] is rendered difficult on account of the weakness of the source which does not easily permit one to carry the absorption measurements through three orders of magnitude as is usually required. However, due to the well defined exponential slope of the absorption curve, a half-value layer determination, instead of the end-point method, can be used to define the β energies present.[7]

Applying the usual method for extrapolation to zero-absorption, the following results, summarized in Table 1, are obtained:

Table 1. Analysis from Absorption Curve

Group	*Half-value layer*	*Energy*	*Activity measured*
I	6 mg/cm²	0.25 Mev	58%
	12 mg/cm²	0.4 Mev	
II	—	—	—
III	195 mg/cm²	3.0 Mev	42%

Taking into account several corrections related to β energy and absorber thickness (forward-scattering, etc.)[8], further improvements for better separation of components could be achieved.

It is probable that, due to the slight upward trend in the absorption curve produced by the forward-scattering effect in the range from 10 to 100 mg/cm² absorber-thickness, the intermediate energy of group II (see Table 2) escapes detection.

The energy of the penetrating component is in good agreement with the result of a similar determination carried out by A. Aron on another filter with evenly distributed fallout.[9]

COMPARISON TEST

To improve the energy determination and discrimination, comparison-methods with radioactive standards (RaE, P^{32}, etc.) have been worked out, making use of the full curve rather than of the end-point alone.[10] Again, the practical difficulty of these methods, for weak sources, lies in their dependence on absorption measurements carried all the way to the end of the range.

A new approach to comparison methods is based on the procedure of comparing the transmission of an unknown and a standard emitter with the same absorber, and of resorting to slope determinations of the curve obtained.[11]

Plotting the logarithm of the transmissions of two absorbers against each other for each absorber yields a straight line the slope of which is characteristic of both the standard source and the unknown emitter. Once a calibration curve for a whole range of known energies is obtained, determination of any unknown sample can be performed quickly with only a small number of absorption measurements. Notwithstanding the great ease and simplicity of this method, energy and relative intensity of components can be obtained with a precision comparable to the more elaborate procedures formerly in use.

On applying this procedure to the particle on the filter, the results of Table 2 have been obtained (see Fig. 5). The percentage distribution of soft and hard components is different from that of Table 1, in consequence of an increased counter-window thickness (3.5 mg/cm²) used for this analysis, as can be shown.

Table 2. Analysis by the Harley-Halden Comparison Method

Group	*Av. slope*	*Av. energy Mev*	*Measured intensity %*
I	3.0	3.1	72
II	0.462	1.12	13
III	0.054	0.3	15

These energies can be attributed, with varying degrees of probability, to the following fission products present:[12]

Table 3. Constitution of Particle

Group	*Isotope*	*Energy Mev*	*Relative group abundance %*
I	$_{45}Rh^{106}$	3.5	10 (probable)
	$_{59}Pr^{144}$	3.2	80
	$_{39}Y^{90}$	2.4	10 (probable)
II	$_{38}Sr^{89}$	1.48	(probable)
	$_{55}Cs^{137}$	1.2	
	$_{38}Sr^{90}$	0.6	(probable)
III	$_{58}Ce^{144}$	0.36	
	$_{55}Cs^{137}$	0.5	
	$_{61}Pm^{147}$	0.2	(probable)

Each of these isotopes has a sufficiently long half-life, either in itself or in its parent, and also a sufficient fission yield to make its presence in the particle possible. Other fission products of similar characteristics, are ruled out on account of strong γ radiation associated with them. The complete lack of α rays precludes also the assumption of any original bomb material present.

Taking into account relative intensity and counting efficiency for the different energy groups, an estimate

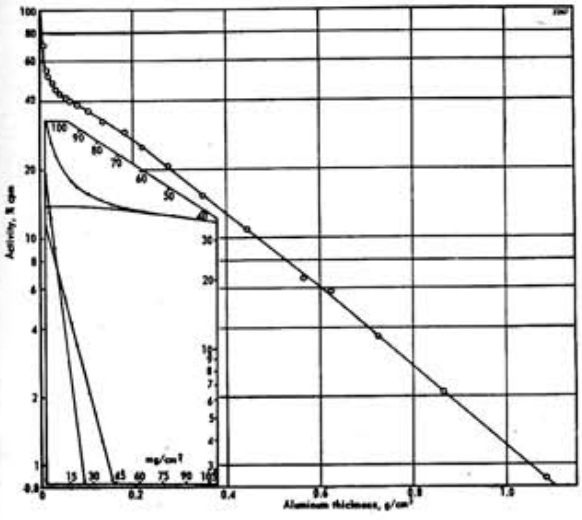

Figure 4. Al absorption curve of radioactive filter-particle after reduction to vertical incidence. Inset: Expanded low-energy part of curve

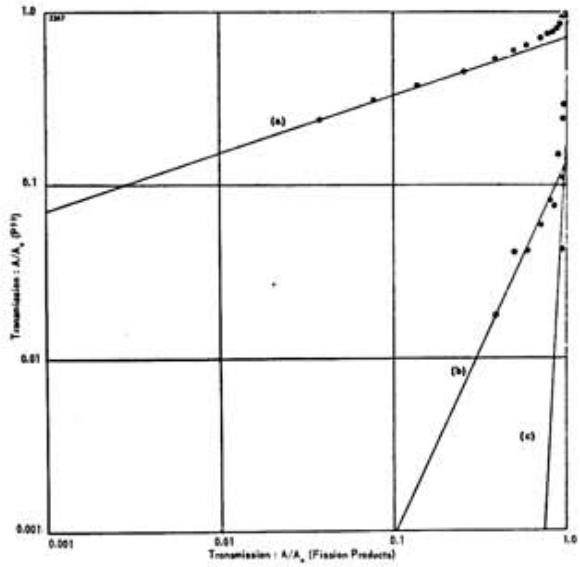

Figure 5. Plot of mixture of fission products versus a P^{32} standard. (*a*) reciprocal slope = 3.0; (*b*) reciprocal slope = 0.462; (*c*) reciprocal slope = 0.054

Table 4. Relative Isotopic Abundance

$_{58}Ce^{144}$	(40%)	→	$_{59}Pr^{144}$	(40%)
$_{38}Sr^{90}$	(5%)	→	$_{39}Y^{90}$	(5%)
$_{45}Rh^{106}$	(5%)	$_{38}Sr^{89}$, $_{55}Cs^{137}$, $_{61}Pm^{147}$		(5%)

(see Table 4) of the relative amount of isotope activity present can be made.

Two chains, therefore, are responsible for the near totality of the activity present.

ABSOLUTE CALIBRATION

Using a calibrated $_{91}Pa^{234}$ source (2.3 Mev) under identical conditions of back-scattering, etc., the absolute activity of the high energy β emitters was determined, on 10 January 1958, as being 1.2×10^{-9} curie (or 44 dps). For the hard β component no additional correction for the absorption in the filter is needed. The result is in agreement with Gross' activity determination of 5×10^{-9} curie, or approximately 2×10^{-9} curie for the hard component (see Table 1), carried out 230 days earlier (half-life of $_{58}Ce^{144}$ = 275 days).

According to Table 4, 32 dps are to be attributed to $_{59}Pr^{144}$ and 4 dps to each of the other hard β emitters present ($_{45}Rh^{106}$, $_{39}Y^{90}$, $_{55}Cs^{137}$). For the number of radioactive atoms, making up the particle, only the much longer-lived parents ($_{58}Ce^{144}$, $_{38}Sr^{90}$, $_{44}Ru^{106}$, and $_{55}Cs^{137}$) are to be taken into account.

Since equilibrium conditions prevail, we have

$$N\ (\text{atoms}) = \frac{dn}{dt}\Big/\lambda_1(\text{Ce}) + \frac{dn}{dt}\Big/\lambda_2(\text{Sr}) + \frac{dn}{dt}\Big/\lambda_3(\text{Ru}) + \frac{dn}{dt}\Big/\lambda_4(\text{Cs}) \quad (5)$$

leading to the value of $N = 11{\cdot}5\times10^9$, or 6.1×10^9 atoms, depending on whether Cs is included or not.

This represents a volume with a radius of 0.5 or 0.4μ respectively, thus confirming the character of a lower limit for Gross' estimate of the volume of the active mass carried out 8 months before. In fact, if one would assume only Ce to be present, $N = 1.4\times10^9$ and $r = 1/4\mu$ result.

Finally, a direct microscopic measurement on the particle itself was performed. As expected, its actual size is considerably bigger (radius = 4μ) than its active mass.

Activity and size, as well as the almost perfect spherical shape of the condensation nucleus, may lead to a better insight into the physical facts underlying formation, transport and subsequent contamination of such nuclei in atomic test explosions. Since, in May and June, the thermal equator lies north of latitude 0°, the particle may be due to tropospheric rather than to stratospheric fallout.

Great fluctuations of fallout activity and particularly strong isolated single values have been measured elsewhere too, as may be seen, for instance, from the reports of the US Naval Research Laboratory in Washington, which contain observations from several South American stations.

The present results may lead one to believe that many of these extraordinary observations must have been caused by single particles of much higher than medium activity.

The customary reduction of fallout activity to unit air volume (cubic meter or cubic foot) would appear unjustified in cases where the activity is predominantly produced by single particles, since for different sampling times there would result different activities. The reduction to unit air volume is only justified, when the number of particles is big enough to guarantee a statistical distribution.

These results also have a bearing on the biological effects of radiation. Sr^{90} and Sr^{89}, chemically similar to Ca, are largely deposited in the calcifying tissue of bone. The rare earths related elements, as Ce^{144} and Y^{91}, are also bone-seekers. The radiations from all these elements held in various parts of the osseous structure can impair blood forming tissues and cause tumor formation, particularly if concentrated in single specks of abnormally high activity.[14]

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APPENDIX I

Self-absorption in Filter[1]

The particle is trapped in the filter at some unknown depth. The absorption of the filter can be found by

measuring the activity while (*a*) the exposed surface of the filter is in direct contact with the counter; (*b*) the unexposed surface of the filter is in direct contact with the counter; and (*c*) an unexposed filter is placed between the exposed surface and the counter.

Taking γ as the absorption coefficient of the filter material and x_1, x_2 as the depth of the particle, respectively, below the exposed and the unexposed surface, the following relations for the three cases above apply:

$$I_1 = I_0 \exp(-\gamma x_1) \quad (a)$$
$$I_2 = I_0 \exp(-\gamma x_2) \quad (b)$$
$$I_3 = I_0 \exp[-\gamma(x_2 + x_1 + x_2)] \quad (c)$$

yielding

$$I_0 = I_1(I_2/I_3)^{\frac{1}{2}}.$$

Introducing the values: $I_1 = 314$ cpm, $I_2 = 268$ cpm, $I_3 = 183$ cpm, one obtains $I_0 = 1.21$ or an absorption of 21% in the filter. This corresponds to an Al-equivalent of 25 mg/cm² (see inset of Fig. 2) and to the Feather range of a 135 kv β ray.

APPENDIX II

Transformation to Parallel Beam Incidence

If $\psi(x)$ is the absorption curve for a parallel beam, $J(x)$ the measured curve, x the thickness of the absorber and θ_0 the maximum value of θ (see inset of Fig. 1), then one has (under the assumption of both primary and secondary radiation being scattered mostly in a forward direction):

$$J(x) = \int_0^{2\pi} d\varphi \int_0^{\theta_0} \psi\left(\frac{x}{\cos\theta}\right) \sin\theta d\theta. \quad (1)$$

In the present case, as experimentally verified, the efficiency of the counting system is practically uniform over the entire aperture of incidence. Therefore, no additional "sensitivity function"[13] needs to be taken into account.

According to Gross (Ref. 4, Eq. (2)), the integral (1) can be transformed into

$$\psi(x) = f(x) + a\psi(x/a) \quad (2)$$

where
$$f(x) = \frac{1}{2\pi}[J(x) - xdJ(x)/dx] \quad (3a)$$

and
$$a = \cos\theta_0 \quad (3b)$$

Being $a < 1$, one can solve (2) by the expansion

$$\psi(x) = \sum_{n=0}^{\infty} a^n \psi_n(x). \quad (4)$$

An iteration method, equalling terms with identical powers of a gives,

$$\psi_n(x) = f(x/a^n). \quad (5)$$

The final solution becomes

$$\psi(x) = \sum_{n=0}^{\infty} a^n f(x/a^n). \quad (6)$$

The first term coincides with the usual form of the Gross transform.

It is frequently convenient to draw the absorption curve as $\log J(x)$.

Then one has:

$$f(x) = \frac{1}{2\pi} J(x)[1 - xd \log J(x)/dx]. \quad (7)$$

It has been found empirically, that the absorption curve for a single β emitter is exponential. If,

$$J(x) = J^0 \exp(-\alpha x) \quad (8)$$

one has

$$f(x) = \frac{1}{2\pi} J(x)[1 + \alpha x]. \quad (9)$$

This curve starts with a horizontal tangent.

REFERENCES

1. A. Aron and B. Gross, Z. Naturforsch, *12a*, 944 (1957).
2. K. Way and E. P. Wigner, Physiol. Rev., *73*, 1318 (1948).
3. H. F. Hunter and N. E. Ballou, Nucleonics, *9* (November 1951).
4. B. Gross, Z. Physik., *83*, 214 (1933).
5. N. Feather, Proc. Cambridge Phil. Soc., *34*, 599 (1938).
6. L. E. Glendenin, Nucleonics, *2* (January 1948).
7. J. M. Cork, *Radioactivity and Nuclear Physics*, D. Van Nostrand Co., New York (1957).
8. B. P. Burtt, Nucleonics, *5* (February 1949).
9. L. R. Zumwalt, Rep. Mon. C-393 (1949).
10. A. Aron, Anais. acad. brasil. cienc, *28*, 423 (1956).
11. L. Katy and A. S. Penfold, Rev. Mod. Phys., *24*, 28 (1952).
12. J. H. Harley and N. Hallden, Nucleonics, *13* (January 1955).
13. W. Jentschke, Z. Physik, *120*, 165 (1943); J. M. Siegel, Rev. Modern Phys. *18*, 513 (1946); Nat. Nuclear Energy Ser., Vol. IV/9, McGraw Hill Co., New York (1951).
14. L. Janossy, Z. Physik, *99*, 369 (1936).