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SEARCH FOR SUPERHEAVY ELEMENTS IN NATURE

E. Cheifetz,† R. C. Jared, E. R. Giusti, and S. G. Thompson

Nuclear Chemistry Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

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ABSTRACT

The results of a search for elements in the region of atomic number 114 are reported. More than 40 large samples of ores, natural minerals and other possible sources have been investigated. The method of detection made use of a large liquid scintillator to detect spontaneous fission events in which unusually large numbers of neutrons are counted. The apparatus was located in a tunnel under about 250 meters of earth shielding in order to reduce the interfering effects of cosmic rays. The limit set for the ratio of half life/concentration for all the materials tested was greater than 10^{23} years.

I. INTRODUCTION

In recent years various estimates and calculations have been made which suggest the possibility that nuclei having atomic numbers in the region 108 - 114 may be sufficiently stable to exist in nature.[1,2,3] These nuclei are expected to decay either directly by spontaneous fission or by alpha and/or electron capture to give products that would undergo spontaneous fission. The production of such long-lived superheavy nuclei in nature could come about via the r-process[4] or by fusion of neutron-rich heavy ions in the vicinity of neutron stars.[5] On the basis of the present knowledge of fission barriers and spontaneous fission decay of heavy elements, the possibility of production of the superheavy elements in nature can neither be excluded nor be established; thus the field is open for experiments aimed at discovering minute quantities of these elements in nature.

Several searches for the presence of superheavy nuclei in nature have been carried out by attempts to detect fission events whose origin cannot be attributed to spontaneous fission of naturally occurring known species such as ^{238}U or attributed to fission induced by cosmic rays and natural neutron fluxes in heavy elements. Tses'lyak[6] has reported the observation of fission tracks in samples of lead glass which he attributed to the possible decay of superheavy elements. Flerov et al.[7] have extended the study of Tses'lyak by measuring actual spontaneous fission events in a large shielded proportional counter in which they placed the lead glass that showed the fission tracks described by Tses'lyak. On the basis of additional observations of a similar rate of fission events in some lead ore samples Flerov et al. concluded that they had indeed found spontaneous fission events with an apparent half life

of 4×10^{20} years relative to the major component of their samples which they attributed to the presence of superheavy elements in nature. On the other hand P. B. Price et al.[8] reported failure to observe accumulation of fission tracks in old lead-rich and gold-rich minerals, thus restricting the spontaneous fission half life to more than 2×10^{23} years relative to the lead and gold concentration of these minerals. The latter authors consider the results of their work to be in contradiction with the remarkable results of Flerov et al.[7]

Estimates by Nix[9] based on liquid drop dynamical calculations and simple extrapolation based on the behavior of the kinetic energy release and the mass differences between the fissioning nuclei and the fragments of a wide variety of nuclei indicate that on the average ~230 MeV of kinetic energy should be liberated in the fission of the superheavy nuclei and that about ten neutrons should be emitted in such fission events. The empirical approach to the estimate of the neutron multiplicity in the spontaneous fission of superheavy elements is presented in appendix A.

The emission of such a large number of neutrons from each single fission event is then a very sensitive indicator of the presence of superheavy elements as this property does not occur in any of the presently known spontaneously fissioning species in nature. Furthermore, these evaporated neutrons would have average energies in the range of 2 - 3 MeV and thus would be able to escape from a large sample and enter a counting chamber. Consequently large amounts of materials can be examined with a sensitivity which is greater by 10^2 - 10^3 fold in comparison with elaborate electronic fission fragment-counting methods which require the effective thickness of the sample to be less than the fragment ranges.

Neutron counting as a method of searching for the presence of decaying superheavy elements has been used by Grimm et al.[10] who have measured many samples and obtained negative results with limits of half life/concentration of 2×10^{19} years.

We have used a large gadolinium loaded liquid scintillator detector as a sensitive method for detecting events in which several neutrons are emitted. The efficiency for detecting each single neutron was high (~65%) and furthermore, due to the wide variation in thermalization and capture time of the neutrons in the detector's liquid, the system was capable of counting the individual neutrons and recording the multiplicity distribution of such events. The system was placed in a 250 m deep tunnel to sharply reduce the background of energetic cosmic rays inducing spallation and fission reactions that involve emission of many neutrons.

In the following paper we summarize the results found in over one and a half years of experimental effort. A preliminary report on this experiment was given at the Leysin conference.[11]

II. DESCRIPTION OF THE METHOD OF DETECTION

The neutrons are detected by a large gadolinium loaded liquid scintillator with the capability of measuring neutrons with high efficiency. A schematic view of the detector system is shown in Fig. 1. Its main body is a tank with dimensions $62 \times 62 \times 125$ cm which holds a liquid consisting of toluene solvent, ~ 8 g/ℓ gadolinium octoate, 0.1 g/ℓ POPOP and 5 g/L p-terphenyl. Sixteen 5-inch Dumont No. 6364 photo-multipliers are mounted on glass viewing ports on two walls of the chamber. The inner walls of the chamber are painted with a white reflective coating. A tube for holding the samples is located

at the center of the chamber and has the dimensions of 11.4 cm diameter and 105 cm. depth.

The system was rebuilt from many components of a system already described in the literature.[12,13] The dimensions of the detector tank were dictated by the entrance to the tunnel in which it is located.

Neutrons produced by any source placed at the center of the chamber enter the liquid and are thermalized by collisions with the hydrogen in the solution and eventually are either captured by the gadolinium or leak out of the tank. The (n,γ) reaction in gadolinium produces ~ 9 MeV of gamma energy, the energy usually being shared by several gamma rays. The electrons created by the reaction of these gamma rays with the liquid produce scintillations which are seen by a few of the photomultipliers.

The distribution in time for neutron capture is broad and has a peak at about 10 μsec after the neutrons are emitted. About 90% of all the captured neutrons are detected within the interval 1-36 μsec after their production. In this way individual members of a burst of energetic neutrons are separated in time for convenient electronic multiplicity counting.

Due to the large volume of samples it is impossible to trigger the system with the fission fragments; therefore the system is designed to be triggered by the first neutron or the prompt gamma rays from any fission event.

III. SOURCES OF BACKGROUND

When the system operates with any neutron or gamma signal as the main trigger, it is sensitive to the environmental radiation which consists of gamma rays from natural sources (e.g., U, Th, and K) and cosmic rays and the products of their reactions in matter. The gamma rays that arise from

natural sources appear in general as single random pulses. Accidental coincidences between these random pulses yield a distribution which is represented by a Poisson probability function.

$$P(N) = \frac{8.64 \times 10^4 \, (CT)^{N+1} \exp(-CT)}{(N!T)} \qquad (1)$$

where P(N) is the number of events observed per day in which N pulses follow a single random trigger appearing at a rate of C/sec within a gate length of T(sec) after each pulse. At the operating gate length of 35 μsec and a count rate of 600 counts/sec less than 0.5 events per day appearing as multiplicity four or more are expected from the above probability function.

The experimental equipment was placed in a 250 m deep (500 meter water equivalent) tunnel to reduce the cosmic-ray background. The situation in regard to cosmic-rays underground was reviewed by E. P. George.[14] At the depth of the tunnel the only significant component of the cosmic rays that can induce multiple neutron emission are energetic μ-mesons, the intensities of which are approximately 1000 times smaller than on the surface of the earth.

At the depth of 600 meter water equivalent (m.w.e.) about 10^{-4}"stars"/cm^3-day have been found following charged particle tracks in a photographic emulsion. "Star" events occur when μ-mesons either stop or are inelastically scattered by a heavy nucleus which then recoils while emitting neutrons. This is equivalent to observing an event with high neutron multiplicity.

Gorshkov and Zabiyakin[15] have studied neutron production induced by μ-mesons at a depth of 150 m.w.e. and estimated that at a depth of 800 m.w.e. the total rate of neutron production in lead is $\sim 10^{-8}$ neutron/g-sec. Less than 10% of the neutrons are produced from stopped negative μ-mesons for which

$\bar{\nu} = 1.8$ in lead whereas 90% of the neutrons are produced from inelastically scattered μ-mesons with $\bar{\nu} > 10$ and thus could register efficiently in our chamber as significant events. Assuming $\bar{\nu} = 10$, we expect a rate of 50 high multiplicity events for a sample of 50 Kg lead in 250 hours.

The passage of μ-mesons in the detector liquid causes an ionization of ~2 MeV/cm of path length, thus a μ-meson entering the sample can be identified by its high energy loss. If a high energy inelastically scattered μ-meson comes into the tank from the vertical direction through the sample tube it must traverse at least 20 cm of the scintillating liquid at the bottom of the tank, depositing more than 30 MeV in the tank.

Since neutron capture in gadolinium gives a total of 9 MeV of gamma ray energy, it was possible to eliminate some of the μ-meson induced high multiplicity events by rejecting the events which produced a signal that was equivalent to ionization of more than ~9 MeV in the liquid scintillating tank. In addition to this a liquid scintillator detector of thickness 12.5 cm was placed above the chamber as shown in Fig. 1 to reject the events induced by vertical μ-mesons. Nevertheless the neutron multiplicities which are induced in the sample by energetic neutrons which are produced by inelastic collisions outside the chamber cannot be rejected nor can the μ-meson induced neutron multiplicities detected in the chamber but originating outside it be eliminated.

IV. ELECTRONICS

A simplified block diagram of the electronics is shown in Fig. 2. The photomultiplier pulses are summed in two banks (8 photomultipliers in each). A "tank pulse" is obtained when coincident pulses are observed from banks A and B and when the linear sum of all photomultiplier outputs corresponds in

pulse height to an energy greater than 1 MeV. This signal has the significance that the light must be seen by at least two photomultipliers and have a sum amplitude corresponding to an energy greater than 1 MeV. Since at least two photomultipliers must give a signal the effect of random noise from individual tubes is minimized. A "tank pulse" triggers, after a 0.5 μsec delay, a 36 μsec gate. (The delay is to ensure that all of the prompt gamma rays from a fission event have been emitted.) During the 35 μsec gate interval all tank pulses are counted by a scaler. At the end of the 36 μsec period the digital information in the scaler is converted to an analog pulse height and transferred to a pulse height analyzer. Then the scaler is reset. The next tank pulse defines a new 36 μsec gate interval.

A spectrum of multiplicity in the range 0-15 is thus obtained in a pulse height analyzer. The system is capable of monitoring a burst of neutrons where the trigger can be activated either by the prompt γ-rays or the first neutron captured. The circuits shown by dotted lines in Fig. 2 are used to remove events that follow detectable cosmic rays.

A signal from either the μ-meson "tank discriminator" or the discriminator of the top cosmic ray detector causes the observed multiplicity to be routed into a separate part of the pulse height analyzer. The "tank discriminator" level which is the level above which a signal from the tank is classified as a μ-meson is set such that it identifies as cosmic rays less than ~1% of the neutrons emitted from a ^{252}Cf spontaneous fission source. Due to the large detection volume and optical attenuation associated with obtaining an appropriate energy response some μ-mesons which reach the sample can perhaps avoid detection.

V. OPERATION OF THE SYSTEM

The efficiency of the system was checked at regular intervals by placing a weak ^{252}Cf spontaneous fission source in the center of the tank and triggering the 35 μsec gate by the fission events (using a small solid-state detector to detect the fragments). The multiplicity distribution obtained in this manner for 25,000 fission events is shown in Fig. 3. Its mean value is $\bar{n} = 2.44$. This yields an efficiency of 65.5% for detection of each neutron (using 3.72 as the average number of neutrons emitted in ^{252}Cf fission).

The pulse height distribution produced by the neutron-induced gamma rays is very wide because of the large size of the chamber and the optical attenuation in the liquid; therefore, the efficiency of the system to detect neutrons depends on the setting of the threshold discriminator. Although the maximum efficiency of the system for neutrons emitted at its center can reach about 72%, an operating efficiency of ~65% was chosen such that the single counting rate due to background gamma rays (with no gamma emitting sample in the tank) was about 600/sec. At this rate about 0.4 events/day of multiplicity four (a trigger followed by four signals within 35 μsec) were recorded due to the gamma background.

Long-term stability and continuous monitoring of the system which is necessary for this type of an experiment was obtained in three ways: 1) The singles counting rate of tank pulses was recorded every 2 sec by a chart recorder and showed the system to be very stable. 2) The data from the multichannel analyzer were printed out automatically at 12 hour intervals and the multiplicity distributions were found to be consistent (within statistical fluctuations). 3) In some experiments photographs were made of the time distribution of the tank pulses that appeared during the 35 μsec gate, whenever an event having a multiplicity of four or more occurred. These events were

checked to be certain they had a normal time distribution and that they did not include electronic noise. The pictures ascertained that the system was operating satisfactorily at the very low counting rates of this experiment. 4) The efficiency of the system was checked twice a week and was generally found to be stable at the ~65% level. Any variation of over 2% in the efficiency and consequently in the single count rate, was corrected by adjusting the threshold discriminator levels. This occurred several times during about one year of running.

An attempt was made to reduce the single count rate of the system by shielding the detector from the natural radiation of the concrete of the tunnel with 5 cm thick steel plates. Although the single gamma-ray rate did decrease from 600/sec to 200/sec at the 65% efficiency level, thus drastically decreasing the random multiplicity rate, the multiplicity rate due to cosmic rays increased significantly either from neutron multiplicities produced by μ-mesons in the 3 tons of iron shielding or by an increase in the production of energetic neutrons which induce secondary multiplicities in the sample. Since there was no physical room in the tunnel for an appropriate shield with low Z (e.g. water), no shielding was used during the actual runs.

VI. RESULTS

Results of the observed multiplicity distribution measured with samples of pure tungsten, lead ore and with an empty chamber are shown in Figs. 4, 5 and 6. The results are plotted on a semilog scale with the abscissa being the multiplicity and the ordinate N! x counts. In this way the multiplicity distribution due to the singles rate, which is the dominant component up to N = 3 appears as a straight line. Linear extrapolation of the straight line

which passes via the points with N = 1, 2 and 3 to the points with N = 4 and N = 5 enables an easy estimate of the contribution there of the random count multiplicities. The slope of the straight line depends on the single gamma ray count rate which for some ore samples was higher than 600 c/sec due to the presence of some gamma emitting natural substances.

The results of many samples that were placed in the detector are summarized in Tables 1-4.

The Tables describe the sample, its weight, and the period of measurement. Under the heading of "Counts" are presented 3 different experimental numbers. The column headed by "4" lists the total observed events with multiplicity of four (i.e. four pulses following a start pulse). The column headed by "4 Ran" is the estimated value of random multiplicities in the "4" column. This value was inferred from linear extrapolation of the counts having multiplicities of 1, 2 and 3. The column "5^+" contains all the observed events having multiplicities of 5 and above. Unless otherwise mentioned the 5^+ events contain negligible random contribution. The "Normalized" column in Tables 1, 2 and 3 presents counts normalized to 50 Kg samples and 250 hour counting periods of multiplicities four or more after subtraction of the "Empty Chamber" count rate and the random multiplicities.

"Normalized" =

$$\frac{[(4)-(4\ \text{Ran})+(5+)]}{\text{Time}} \times 250 - \frac{\text{Empty chamber counts} \times 250}{\text{Time}} \times \frac{50}{\text{Weight}} \qquad (2)$$

The "Normalized" result thus represents the additional counts of multiplicity 4 or more that are due to the presence of the sample. This activity could be caused either by cosmic rays or spontaneous neutron emission.

The value of $T_{1/2}$ /conc. represents the apparent half life of the major component of the sample assuming that all the events that are not accounted for by random multiplicities and empty chamber counts are due to the presence of spontaneous neutron high multiplicity sources. These events also include μ-meson induced events.

Table 1 presents results obtained with processed samples. The "empty chamber" values were measured at three separated periods of time during 18 months of data taking. The pure tungsten, mercury and platinum samples were reagent grade. The lead and copper samples were industrial grade. Since Samples 1-4 of Table 1 are all heavy elements and give rise to similar counting rates in excess of the random events it is plausible to conclude that this activity is due to neutron multiplicities induced by μ-mesons or the products of their reactions. The value of 20 counts/50 Kg - 250h is roughly 10-50% of the expected count rate from the μ-meson induced multiplicities at the 500 m.w.e. depth of the tunnel and is thus taken as the background count rate of the detector for heavy elements from the μ-meson induced neutron multiplicities that escape identification as events induced by μ-mesons.

Samples 9 and 10 of Table 1 were recently processed reagent grade uranium metal which was thus free of the long-lived gamma-ray emitting daughters of uranium. Sample No. 9 was shielded in a 2.5 cm lead capsule to partially absorb the prompt fission gamma-rays and thus prevent them from triggering the system, therefore there were fewer 4+ events with this sample compared with the unshielded sample No. 10. The apparent half life of uranium derived from the observed 4+ events is about 10^{18} y implying that depending on the amount of absorption of prompt fission gamma rays in the sample between 0.7% and 1.5% of all the uranium spontaneous fission events, appeared as 4+ events (this effect will be discussed further in section VI). On the basis

of the sensitivity of the counting system to uranium it is found that $\simeq$ 10 ppm of uranium produces events approximately equivalent to the background rate due to cosmic rays in our samples.

Table 2 is a summary of results obtained on various samples. These samples in general weighed much less than the pure metals and thus we were required to use a large multiplication factor when normalizing to 50 Kg. In these cases there are large statistical uncertainties. The values of $T_{1/2}$/conc. are all about 10^{23} y and all such results are considered to be negative.

In choosing the ore samples attempts were made to select ores which had undergone mechanical concentration but had not been altered by chemical extractions. The mercury sample No. 11 is cinnabar ore obtained from the Great Western Mine, Middletown, California, where it occurs in a highly mineralized Franciscan Formation. The ore had been crushed to about 10 mesh and had been concentrated in several stages up to 80% cinnabar. The age of the ore deposit is about 10^7 years.

A sample of gold nuggets (No. 12) was obtained from the collection of Sierra County, California. The nuggets ranged from thumb to fist-size and were $\approx$ 95% gold. The original gold and quartz bearing veins were probably formed in the late Jurassic period (1.5×10^8 years ago).

High grade galena ore was obtained from the Bunker Hill mines in Idaho where it occurs in precambrian quartzites and slates. Detailed measurements with these ores and their derivatives also resulted in negative results as discussed in a later section.

High grade platinum ores (sample 13) were obtained as a loan from the Goodnews Bay Mining Company of Alaska. The platinum occurs in small tributaries that cut the eastern flank of the Red Mountains. It comes from weathering

of Dunites that are probably late Paleozoic in age (4-5 x 10^8 years). The ore is a magmatic concentration of platinum minerals in the Dunites. It was further concentrated by a dredge and on mechanical concentration tables to an average of 80-85% of platinum minerals (67% platinum, 2% osmium, 13% iridium, 0.2% ruthenium, 0.1% rhodium, 0.34% palladium, 4.4% gold).

The bismuth sample (No. 14) was collected from many different places in order to get enough material to fill the chamber. Native bismuth, bismuthinite, and bismutite were originally from Colorado, Tanzania, Saxony, Cornwall, Australia and Bolivia. The samples were obtained from Minerals Unlimited, California, Colorado Geological Industries, Colorado, and the Smithsonian Institute, Washington, D.C. The large number of counts for this combined sample was influenced by some of the ore samples containing as much as ~150 ppm uranium.

Sample No. 15 is manganese nodules brought from the ocean floor. The sample was collected from the iron, manganese crust that is 2.5 cm thick, at a depth of 2.74 km at a position 136 km west of the Mid-Atlantic ridge at 45° 31' N, 29° 34' W. This crust started depositing about 16.0 ± 0.8 million years ago and is still being deposited. The sample could have been a host to heavy elements that might have co-precipitated along with the iron and manganese from the ocean waters. This material was used previously in a search for magnetic monopoles in deep ocean deposits.[16]

A large sample of moon rocks and dust brought back on the first United States flight to the moon was also measured (sample No. 16). The samples were probably 2 to 3 x 10^9 years in age. Superheavy elements could have been present in this sample in the ancient moon rocks which have not been altered since the

formation of the moon. Also there could have been superheavy elements formed on the surface of the moon by cosmic particles reacting with the elements in the rocks. Furthermore, superheavy elements could have been produced in outer space and reached the moon's outer layer as part of the cosmic dust.

The copper ore (sample No. 17) was collected from the South Pacific area and loaned to us by the Marcona Corporation. Samples No. 18 and 19 were chosen on the basis of the assumption that superheavy elements might be arriving on the earths surface as cosmic rays. These samples were respectively: activated charcoal and sand that had been used to filter approximately 1100 gallons of drinking water, and an air filter that had been flown by a plane at ~50,000 feet of elevation for ~1 hour (equivalent to ~29,000 $ft.^3$ of filtered air). Sample No. 20 was soil from a pasture in Martinez, California.

Samples 21 - 23 were chosen to check the hypothesis that superheavy elements might not have separated out of the liquid magna chambers in the earths crust and would be concentrated in the last liquid phases along with other elements that have large ionic radii. The rocks selected were pegmatites that appear in dike formations in the Sierra Nevada Mountains of California at Tioga Pass and Tenaya Lake. A composite mixture of the dike rock was selected and counted as sample 21.

A chemical concentration was made on the pegmatites to obtain samples 22 and 23. The chemical procedure used was to grind the rock to about 100 mesh and then to leach the rock with a mixture of 2 parts agua regia and 1 part water. The resulting solution of sand and acid was stirred four times during a period of not less than 12 hours (temperature 20 - 25° C.). At the end of the leaching periods the acid mixture was decanted off and two water rinses

followed. The resulting liquid was then evaporated to near dryness so that the volume could be fitted into the counting chamber. Of the material dissolved from the rock 66% was present in the sample counted.

Sample 22 consisted of leached material from 338 Kg. of pegmatite rock from the east end of Tenaya Lake, California, and sample 23 was leached material from 122 Kg. of rock that came from Tioga Pass, California. The leaching procedure dissolved substantial amounts of natural radioactive material from the rock so that the singles background counting rate for samples 22 and 23 was ~1800 counts per second. This is to be compared with a rate of 600 counts/sec for normal samples. This background accounted for all of the multiplicity 4 events which were observed in these cases. No attempt was made to normalize the results obtained on these samples because of the large uncertainties involved in the leaching efficiency for superheavy elements.

Special attention was paid to the investigation of lead ores since element 114 is expected to be a homologue of lead. As mentioned before, Flerov et al.[7] have reported finding statistically significant spontaneous fission activity with $T_{1/2}$/conc. of $\sim 5 \times 10^{20}$ y in several lead samples. The first lead ore sample that was checked (sample No. 24) showed significant excess neutron activity above the normal level obtained with the samples. A second lead sample (No. 25) was then prepared from the same ore body. The second sample was prepared at the U.C. Richmond field station (about 5 miles away from the LBL main center) by persons who had minimum contact with spontaneous fission sources in order to minimize possible contamination.

Both samples were prepared from high grade galena ore that had been obtained by the Materials Science and Engineering department of the University

of California at Berkeley about 30 years ago from the Bunker Hill mines in Idaho. The ore occurs in precambrian quartzites and slates and had probably been deposited 7.5×10^7 years ago. The ore was crushed to less than 1/2 inch mesh size and then was hand-sorted to remove as much of the impurities as possible. In this way a composition of ~80% galena was obtained.

The second sample (No. 25) also showed a high activity similar to the previous sample. From the ratio of the counts at multiplicity 4 and 5^+ it was inferred that $\bar{\nu} = 2.5\text{-}5$. The samples were then checked for uranium concentration by two independent methods: 1) neutron activation with observation of fission tracks in mica sheets placed in contact with the irradiated samples, 2) measurements of gamma ray spectra in order to detect uranium daughters. Both methods showed that the uranium concentration in the ore was less than 0.5 ppm and thus could give rise to less than 2% of the observed activity. The mica study showed that the uranium was concentrated in the rock present with the ore and not in the galena itself. Therefore sample No. 28 which contained mostly rocks along with about 30% galena compared with ~80% galena in samples No. 24 and No. 25 was prepared. This sample did not show excess neutron activity, the result indicating at this stage that the activity originated from the galena.

In the next stage an attempt was made to find out whether excess neutron activity concentrates in any of the fractions separated in the processes of a lead smelting and refining plant. Nine samples were collected at the Selby plant in Crockett, California. The samples were taken from most of the locations at which chemical fractionation occurs at the plant. No significant activity was found in any of these samples as is shown in Table 4. Each of the nine samples had a volume of about 5 liters. This was the same volume as

that of the lead ore samples which gave significant neutron activity. If any concentration of activity occurred in the processing, the activity measured with a 5 liter sample in 250 hours should have exceeded the activity from the same volume of lead ore of about 35 counts/250 hours. Such a result was not observed.

Following the negative results on fractions from the Selby plant, lead ore and lead bullion were obtained from the Bunker Hill mines in Idaho from which location the lead ore in our possession was mined ~30 years ago. Both samples also gave negative results. Then the attempt was made to concentrate the activity in ~200 Kg of the high grade lead ore that was at our disposal. The process chosen for the separation was reduction of the galena by iron in the reaction $PbS+Fe \rightarrow Pb+FeS$. The lead ore was crushed into a fine powder and mixed with 5.7% Sodium Borate and 18.0% iron dust. Powdered carbon was placed on top of the mixture to prevent oxidation of the lead. The mixture was put into covered stainless steel cans which were placed in an oven at 1100° C. for about 3 hours. The sodium borate becomes a liquid at 741° C. and thus helps bring about contact between the crushed galena and the iron in order to improve the efficiency of the reaction.

The contents of the steel cans separated easily into three components: 1. lead bullion, 2. slag with iron, 3. slag with sodium borate. The three components were poured into separate sets of containers, each set with a volume of about 7 liters. The slag 1 and slag 2 sets of containers included only about 50% of the total slag produced. The lead bullion represented a concentration in volume by a factor of 8 as compared with the lead content of the previously measured lead ore. The slag represented concentration by a factor

of 4. The samples were measured and all showed insignificant neutron activity compared with the expected activity on the basis of the concentration factors.

Before undertaking steps to trap the vapor that might have escaped in the separation process, two more samples from the same lead ore body were measured, (No. 29 and No. 30). These samples gave negative results whereas the original lead ore samples that appeared positive were already used in the separation process.

The long search into neutron activity in lead ores was thus concluded with negative results as no concentration of the apparent activity was achieved and the observed significant activity could not be reproduced by other samples from the same ore. It is possible that the activity could have been due to very slight contamination by ^{252}Cf that could have occurred despite the strict precautionary measures.

VII. DISCUSSION

The results of the previous section were based on the observation of multiplicities of four or more neutrons following a triggering of the system by a neutron or gamma ray and the assumption that all spontaneous fission events of superheavy elements would emit such observable multiplicities.

The results must be modified if the spontaneous fission events give a different distribution of 4^+ events. This is the case with ^{238}U where only ~1% of the fissions were detected as 4^+ events. The sensitivity for detection of the above mentioned multiplicities depends on the shape of the neutron probability distribution. Studies of the distribution of neutrons were carried out by several authors[17, 18] for spontaneously fissioning isotopes of U, Pu, Cm, and Cf. The shape of the neutron distributions was fitted successfully

by the equation:

$$P(\nu) = \frac{\nu_{max}!}{\nu!\,(\nu_{max}-\nu)!} \left(\frac{\bar{\nu}}{\nu_{max}}\right)^{\nu} \left(1-\frac{\bar{\nu}}{\nu_{max}}\right)^{\nu_{max}-\nu} \qquad (3)$$

where the condition $\nu_{max} = 2\,\bar{\nu}$ is imposed.

The form of the neutron multiplicity distribution in spontaneous fission is associated with the variation in the kinetic energy of the fragments and with the statistical nature of the neutron evaporation process. The probability for detection of a 4+ event (1 trigger neutron + 4 or more neutrons) is

$$\text{Probability of } 4+ = \sum_{\nu=5}^{\nu=\nu_{max}} \sum_{N=5}^{N=n} P(\nu)\epsilon^{N}(1-\epsilon)^{\nu-N} \binom{\nu}{N} \qquad (4)$$

The results are shown in Fig. 7. Two experimentally measured points are shown on the curve. The ^{252}Cf point was measured by using a fission trigger, and the ^{238}U point was obtained by placing 10 grams of ^{238}U metal in the chamber without shielding. In the latter case the result is that some of the prompt gamma rays trigger the gate and thus more 4^+ events were observed than would have been expected on the basis of neutrons alone.

The validity of the extrapolation of $P(\nu)$ to high $\bar{\nu}$ values becomes less critical with increase of $\bar{\nu}$ because a larger proportion of the fission events result in observed 4^+ events and the results depend less on a correct description of the upper tail of the $P(\nu)$ distribution. The calculation does not include the effect of the prompt gamma rays as most of them are absorbed within the bulky samples. The prompt fission gamma rays can reduce the required neutron multiplicities from 5 to 4 and Fig. 7 indicates a lower limit of the sensitivity of the system. The effect of dead time (0.2 μsec) after each pulse is not included in the calculation. This reduces the probability to

to detect a 4^+ event by ~10% which is perhaps compensated for by the neglect of the prompt gamma ray effect.

An indication of the shape of the multiplicity distribution can be obtained from the ratio of the 4 events (events of multiplicity 4) to the 5^+ events along with the assumption that $P(\nu)$ behaves according to Equation 3. In none of the cases did the 5^+ events exceed the 4 events thus indicating that the observed multiplicities all originated from cases with $\nu \leq 6$. If $\bar{\nu}$ exceeds 6 the ratio of $5^+/4$ events according to Equation 4 is of the order of 1. The system attains its maximum sensitivity for values $\bar{\nu} = 7\text{-}13$. For such values of $\bar{\nu}$ it discriminates against spontaneous fission events originating from uranium contamination by a factor of about 100. If the value of $\bar{\nu}$ for superheavy elements is lower than 7 the lower limits given for the existence of superheavy elements must be revised upward in accordance with Fig. 7.

In conclusion, we have been unable to find any indication of the existence of superheavy elements in any of the samples examined; the lower limit of $T_{1/2}$/conc. is ~10^{23} years. The samples counted were chosen to cover a wide range of possibilities as regards concentration of superheavy elements following the pattern of the concentration in nature and artificial processes of the heavy elements with $Z \geq 76$.

The system described here has the advantage of great sensitivity for recording actual fission events with large neutron multiplicities and in addition it could have given direct information concerning multiplicity distributions.

The limit $T_{1/2}$/conc. of 10^{23} years is based on the assumption that the average number of neutrons per fission would be 10 ± 3. Had superheavy elements been detected in any of the samples their half lives must have exceeded about

10^9 years in order to survive since their creation during primordial times. For such a half life the upper limit on the concentration in the samples is thus 10^{-14}.

APPENDIX A. ESTIMATE OF THE NEUTRON MULTIPLICITY

The number of neutrons emitted in spontaneous fission, $\bar{\nu}$, is directly related to the excitation energy of the fragments. Nix[9] has already predicted, on the basis of dynamical liquid-drop calculations and the liquid-drop mass formula that $\bar{\nu} = 10.5$ for the spontaneous fission of the superheavy nucleus $^{298}114$. We shall summarize in the following discussion the empirical evidence that supports this prediction. The average excitation energy of the two fragments can be written as

$$\bar{E}_x = (\bar{\nu} + 1)(\bar{B}_n + \bar{E}_n), \tag{5}$$

where $\bar{B}_n$ and $\bar{E}_n$ are the average neutron binding energies of the neutrons and their average kinetic energies in the center of mass of the moving fragments, respectively. The excitation energy released in the form of gamma rays is already included in Eq. (5), where it is taken to be $\bar{E}_n + \bar{B}_n$ this represents the average gamma emission of a statistical decay of the two excited fragments and $\bar{E}_n$ is approximately equal to the additional gamma-ray energy due to the angular momentum effects in the de-excitation of the fragments. The dependence on $\bar{\nu}$ of the average neutron kinetic energy in the center of mass of the moving fragments has the form found in nuclear evaporation processes and thus can be represented by $\bar{E}_n = 0.75 + 065\ (\bar{\nu}+1)^{\frac{1}{2}}$ (in MeV).[19] The constants in the equation were chosen to reproduce the known results for ^{252}Cf and thermal neutron fission of ^{235}U.

The equation $\bar{E}_x = \Delta\bar{M} - \bar{E}_K$ represents the energy balance of spontaneous fission where $\Delta\bar{M}$ is the mass difference between the mass of the fissioning nucleus and the average mass of fragments and $\bar{E}_K$ is the average kinetic energy of the fragments before neutron evaporation. The neutron myltiplicity expected in the spontaneous fission of superheavy nuclei can thus be derived from an extension of the known systematics of the kinetic energies of fission fragments and from extrapolations of the mass formula. The measured average kinetic energies of the fragments $\bar{E}_K$ (before neutron evaporation) of a wide variety of nuclei has been fitted by Viola and Sikkeland[20] to the equation $\bar{E}_K = 0.1065\ Z^2/A^{1/3} + 20.1$ where $\bar{E}_K$ is in MeV and Z and A are the atomic and mass numbers of the compound nucleus undergoing fission. The above equation applies successfully to fission induced by energetic projectiles as well as to spontaneous fission. Apparently the heaviest compound nucleus for which $\bar{E}_K$ has been measured is $^{278}110$ produced by bombarding ^{238}U with ^{40}A and has been reported by Sikkeland[21] to be 225 ± 4 MeV. The latter result is actually a test of the extrapolation of the empirical fitted relationship which was originally derived when the heaviest known fissioning compound nucleus was ^{254}Fm. The relationship predicts $\bar{E}_K$ to be 217 MeV for the compound nucleus $^{278}110$ for which $Z^2/A^{1/3}=1919$. For the nucleus $^{298}114$ $\bar{E}_K$ is predicted to be 227 MeV ($Z^2/A^{1/3} = 1949$). An alternative way to estimate the kinetic energy is via the liquid-drop calculations of Nix.[22] The calculations do not have adjustable parameters and generally give E_K to be a few percent lower than the experimental values found for most nuclei; however, the dependence on $Z^2/A^{1/3}$ is essentially the same as that of the empirical relationship.

The value of $\Delta\bar{M}$ can be obtained from any of the various mass formulas.

The expected island of stable superheavy nuclei around Z = 114, A = 298 lies along the beta-stability line. The mass of $^{298}114$ is obtained from the smooth part of the droplet-model mass formula of Myers and Swiatecki[23] with the addition of the shell correction of Nilsson et al.[23] The mass of the fragments is taken from the Myers and Swiatecki tables. For a symmetric binary fission ΔM = 308 MeV, whereas for asymmetric fission into Z = 52 and Z = 62 with constant Z/A of the isotope $^{298}114$, ΔM is essentially the same 313 MeV. It is important to note that the stability of superheavy nuclei depends largely on the shell correction to the fission barrier. A 2 MeV variation in the shell correction may have drastic effects on the half lives for decay of superheavy nuclei but would contribute insignificantly to the energy release in the fission of such nuclei. Taking ΔM = 310 MeV and $\bar{E}_K$ = 227 MeV we obtain $\bar{E}_x$ = 83 MeV. The average neutron binding energy for the neutron-rich products is estimated to be 5.5 MeV, thus $\bar{\nu} \approx 10$ with $\bar{E}_n \approx 2.6$ MeV.

Even if the kinetic energy of superheavy nuclei were to vary by 25 MeV from the extrapolated value, $\bar{\nu}$ would still be ≈ 6.5 which is a large value compared to the known values of $\bar{\nu}$ for other spontaneously fissioning nuclei.[24]

The underlying assumption in this discussion is that superheavy elements will undergo binary division. The experimental evidence from fission of uranium induced by ^{40}A indicates that binary division is the predominant mode of fission of the compound nucleus, which was apparently produced.[25] If triple or quadruple fission were to take place there would be a larger energy release, but not necessarily in the form of evaporated neutrons.

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FOOTNOTE AND REFERENCES

† Present address Weizmann Institute of Science, Rehovoth, Israel.

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Table 1. Metallic Samples and Empty Chamber Results

	Sample	Weight Kg	Time hours	Counts 4	Counts 4 Ran	Counts 5^+	Normalized Counts/50Kg-250h	$T_{1/2}$/conc. 10^{22}
1	W	55	233	18	4	10	20 ± 6	16
2	Hg	45	189	13	2	5	20 ± 5	15
3	Pb	91	91	2	1	5	9 ± 5	32
4	Pt	16.8	163.5	6	2	4	25 ± 9	12
5	Cu	60	192	8	3	0	2 ± 4	50
6	Empty		338	6	6	2	1.5 ± 2	
7	Empty		168	7	3	2	9 ± 5	
8	Empty		338	19	13	1	5 ± 4	
6-8	Total		844	32	22	5	4 ± 2	
9	U[b]	0.0107	21.5	29	1	2	1.66×10^6	1.5×10^{-4}
10	U	0.0106	30.5	81	1	9	3.44×10^6	0.7×10^{-4}

[a]Empty chamber "Normalized" column is normalized to 250h counting time and not by weight.

[b]The uranium in sample 9 was shielded by 1" of lead capsule inside the chamber.

Table 2. Results on Ores and Other Samples

	Sample	Weight Kg	Time hours	Counts 4	Counts 4 Ran	Counts 5^+	Mormalized Counts/50Kg-250h	$T_{1/2}$/conc. 10^{22} y
11	Hg Ore	21.2	251	16	4	2	24 ± 12	12
12	Gold nuggets	36	335	21	5	3	11 ± 6	28
13	Pt Ore	20	380	19	10	5	13 ± 7	24
14	Bi Ore	14.3	360	57	21	6	84 ± 24	3
15	Manganese nodules	7	193	23	9	2	120 ± 57	9
16	Moon rocks	3	781	44	18	8	99 ± 58	20
	Steel cans	27						
17	Cu ore	10	279	9	8	1	-20 ± 22	
18	Water filters	10	118	8	4	0	23 ± 22	60
19	Air filters		90	5	2	0		
20	California soil	9	170	15	4	2	83 ± 25	
21	Pegmatite	13.5	310	31	19	4	26 ± 26	
22	Tenaya Pegmatite	<337	135	237	237	4		
23	Tioga Pegmatite	<122	132	570	570	11		

Table 3. Lead Ores and Fractions from Lead Processing

	Sample	Weight Kg	Time hours	Counts 4	Counts 4 Ran	Counts 5^+	Normalized Counts/50Kg-250h	$T_{1/2}$/conc. 10^{22}
24	Galena 1	20	256	35	3	7	86 ± 18	3.4
25	Galena 2	23	77	12	2	3	92 ± 28	3.2
26	Bunker Hill Galena	25	122	6	2	1	12 ± 12	24
27	Bunker Hill Bullion	90	120	14	3	6	17 ± 5	17
28	Low grade Galena	15	46	5	1	0	45 ± 36	6.5
29	Homemade lead [a]	75	240	13	5	4	6 ± 3	48
30	Slag 1 50% [a]	34	70	5	1	0	15 ± 13	73
31	Slag 2 50% [a]	25	185	11	2	5	28 ± 12	54
32	Galena 3	17	112	3	1	0	2 ± 13	145
33	Galena 4	17	94	4	1	1	20 ± 20	15
34	Thalium fraction	7	188	18	5	1	104 ± 50	6

[a]These samples were produced from 135 Kg of lead ore (see text). Samples 30 and 31 are only 50% of the slag produced.

Table 4. Selby Lead Smelting and Refining Plant

Number	Sample	Weight Kg	Time hours	Counts 4	Counts 4 Ran	Counts 5^+	Normalized counts/250h
35	Lead bullion	50	48	2	1	2	12 ± 10
36	Blast furnace slag	19.5	76	8	4	3	19 ± 11
37	Cintering dust (I)	16.1	65	1	2	1	4 ± 8
38	Cintering dust (II)	7.2	108	7	3	0	12 ± 5
39	Blast furnace slag II	12.5	87	1	1	0	-1 ± 3
40	Dust from As,Sb, Sn, Cu removal	5	114	10	3	3	18 ± 8
41	Dust of tin dross	13.1	72	5	2	0	6 ± 6
42	Bismuth fraction	55	74	4	1	2	13 ± 8

Fig. 1. (XBL 7111-4725) The gadolinium loaded liquid scintillator detection system.

Fig. 2. (XBL 706-3251) The electronic system. The dashed lines describe the portion of the system that was used to identify the μ-meson induced multiplicities.

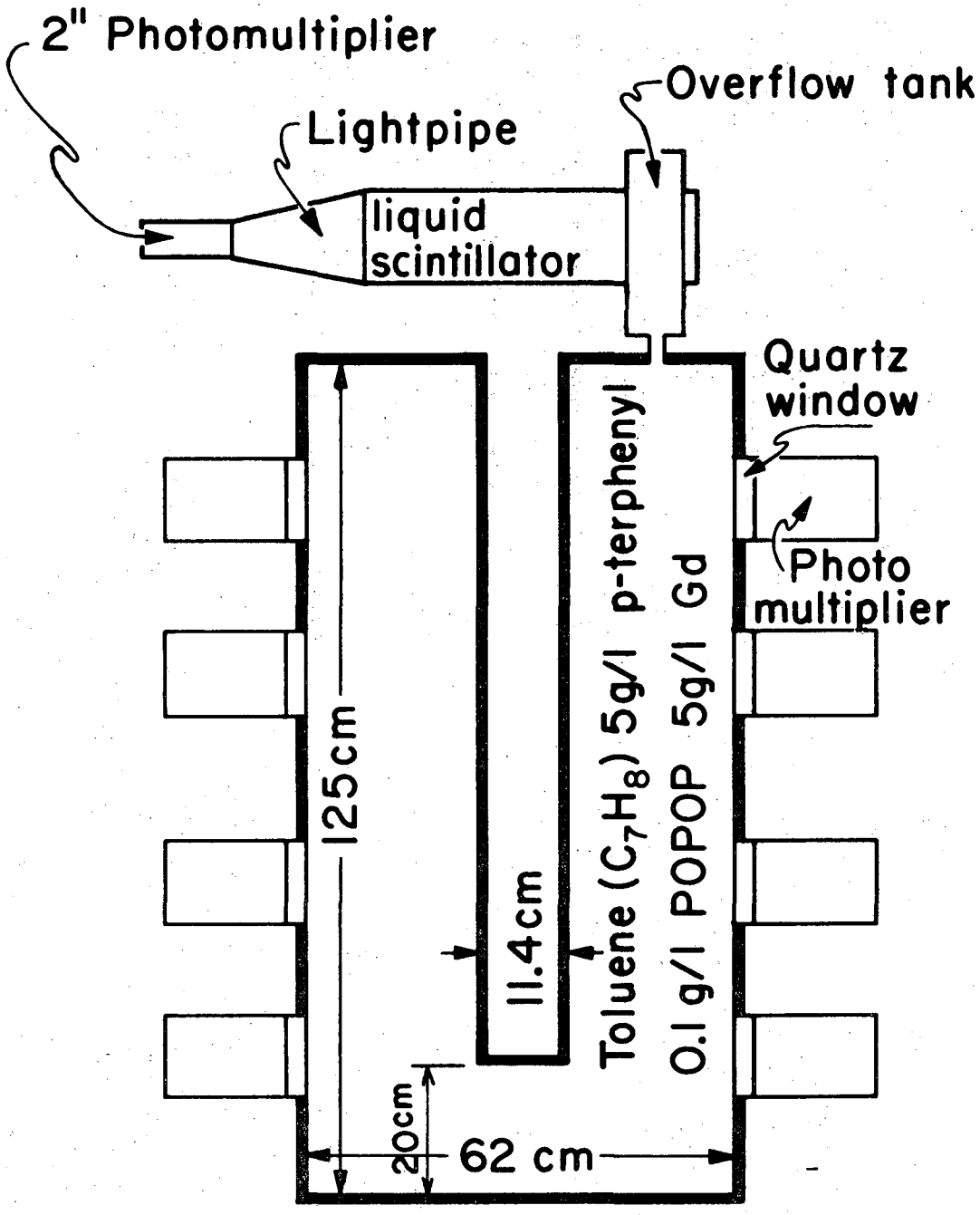
Fig. 3. (XBL 706-3249) The multiplicity distribution observed with a ^{252}Cf source placed in the center of the chambers. The 35 μsec gate was triggered by a fission fragment detector. The average multiplicity of this case was 2.44 i.e., the efficiency ϵ is $\epsilon = 2.44/\bar{\nu} = 65.5\%$; ($\bar{\nu} = 3.72$).

Fig. 4. (XBL 714-3245) The multiplicity distribution for metallic tungsten sample. The abscissa N is the observed multiplicity (i.e. number of events following a trigger). The ordinate is N! x counts of the N-th multiplicity. The straight line represents the contribution of random multiplicities.

Fig. 5. (XBL 714-3244) The multiplicity distribution of the detector without any sample.

Fig. 6. (XBL 7111-4724) The multiplicity distribution of the detector with a lead ore sample.

Fig. 7. (XBL 714-3243) Calculated sensitivity of the system to detect fission events by detecting four neutrons following one neutron which triggers the system, as a function of $\bar{\nu}$. The shape of (ν) is assumed to have the form of equation 3.



XBL7111-4725

Fig. 1

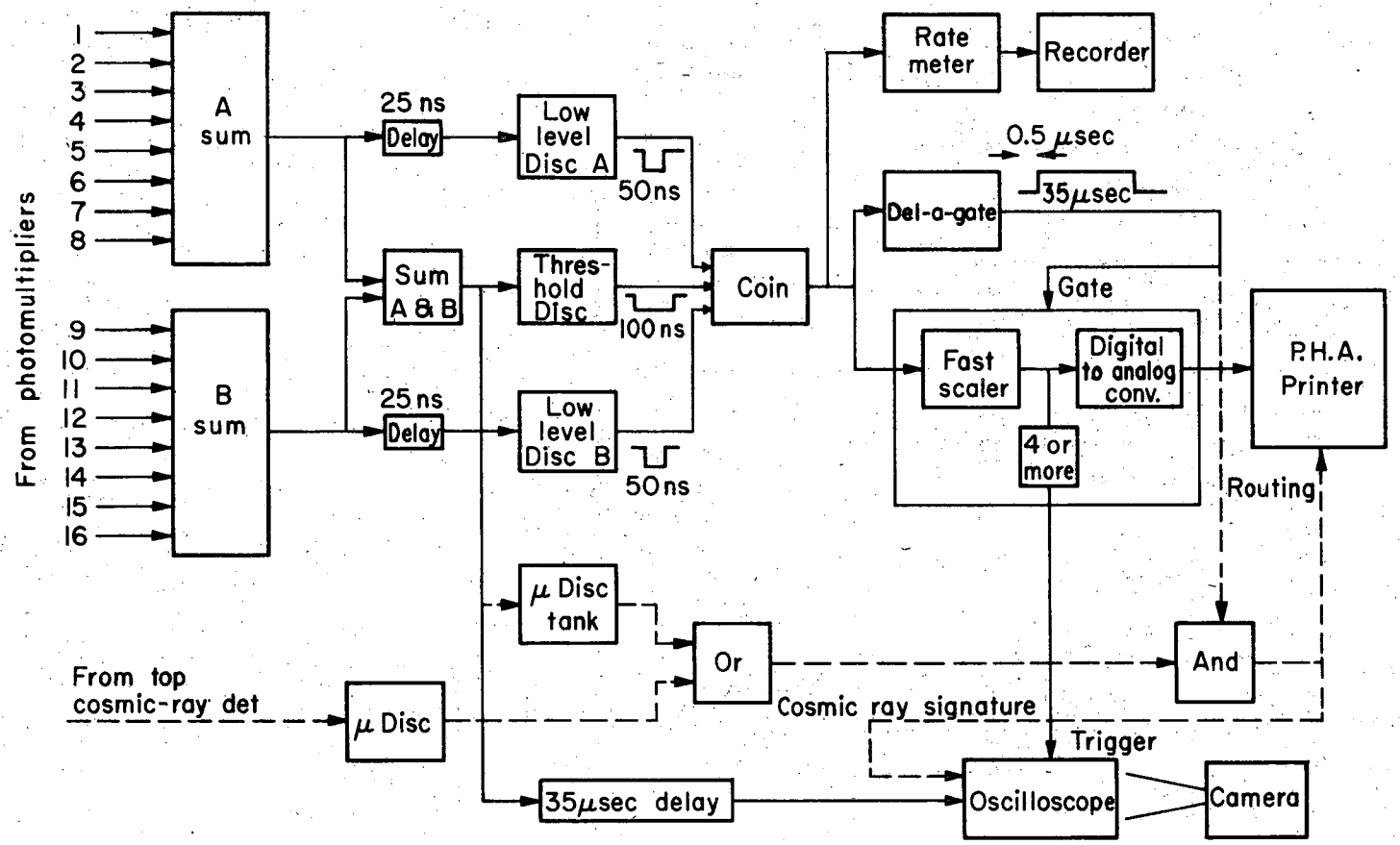
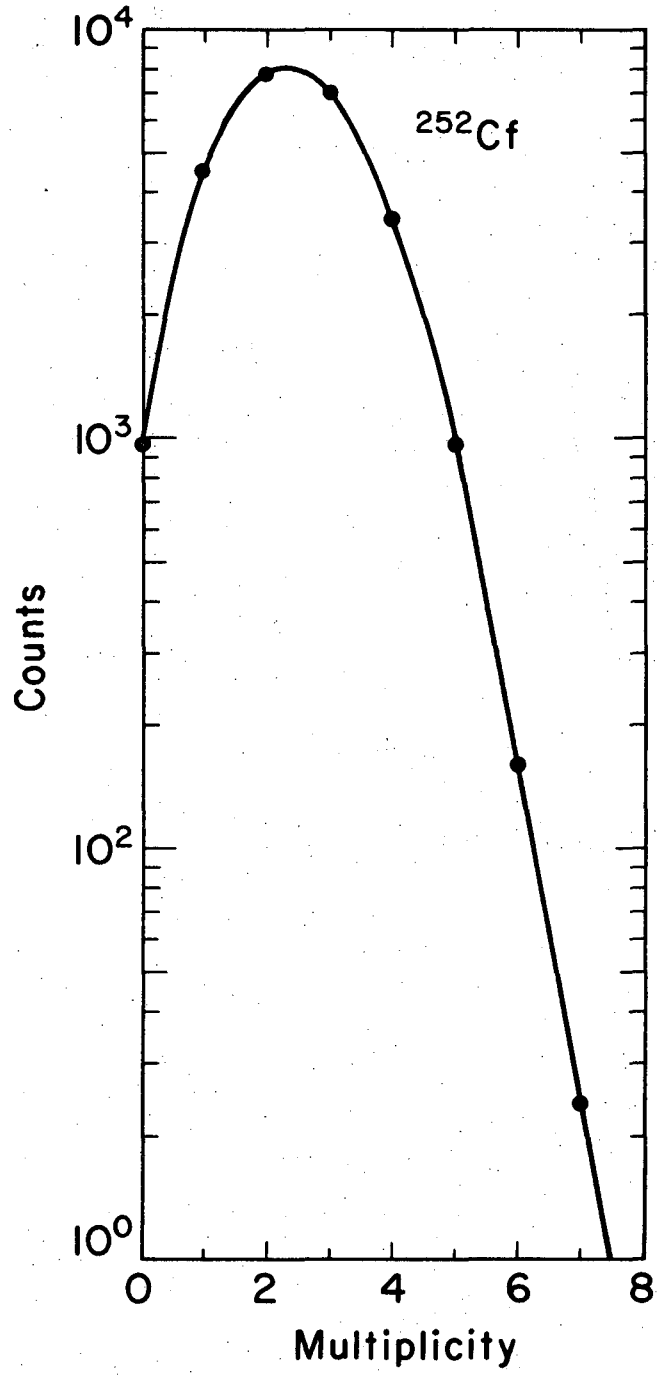

From photomultipliers
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
A sum
B sum
25 ns
Delay
Sum A & B
25 ns
Delay
Low level Disc A
50ns
Thres-hold Disc
100 ns
Low level Disc B
50ns
Coin
Rate meter
Recorder
0.5 μsec
35μsec
Del-a-gate
Gate
Fast scaler
Digital to analog conv.
4 or more
P.H.A. Printer
Routing
μ Disc tank
Or
And
From top cosmic-ray det
μ Disc
Cosmic ray signature
Trigger
35μsec delay
Oscilloscope
Camera
XBL706-3251

Fig. 2



XBL 706-3249

Fig. 3

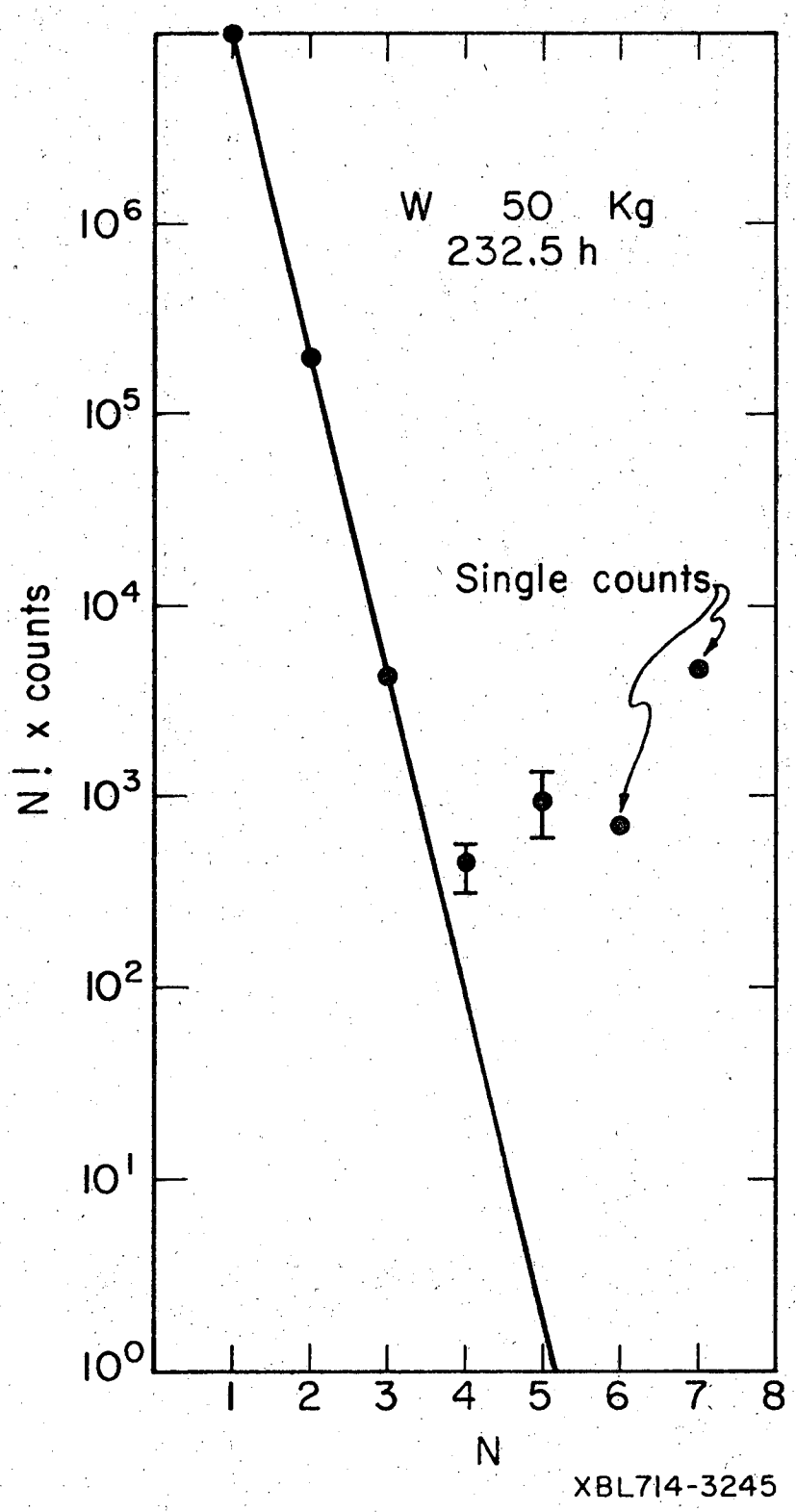

W 50 Kg
232.5 h
Single counts
N! x counts
10^6
10^5
10^4
10^3
10^2
10^1
10^0
1 2 3 4 5 6 7 8
N
XBL714-3245

Fig. 4

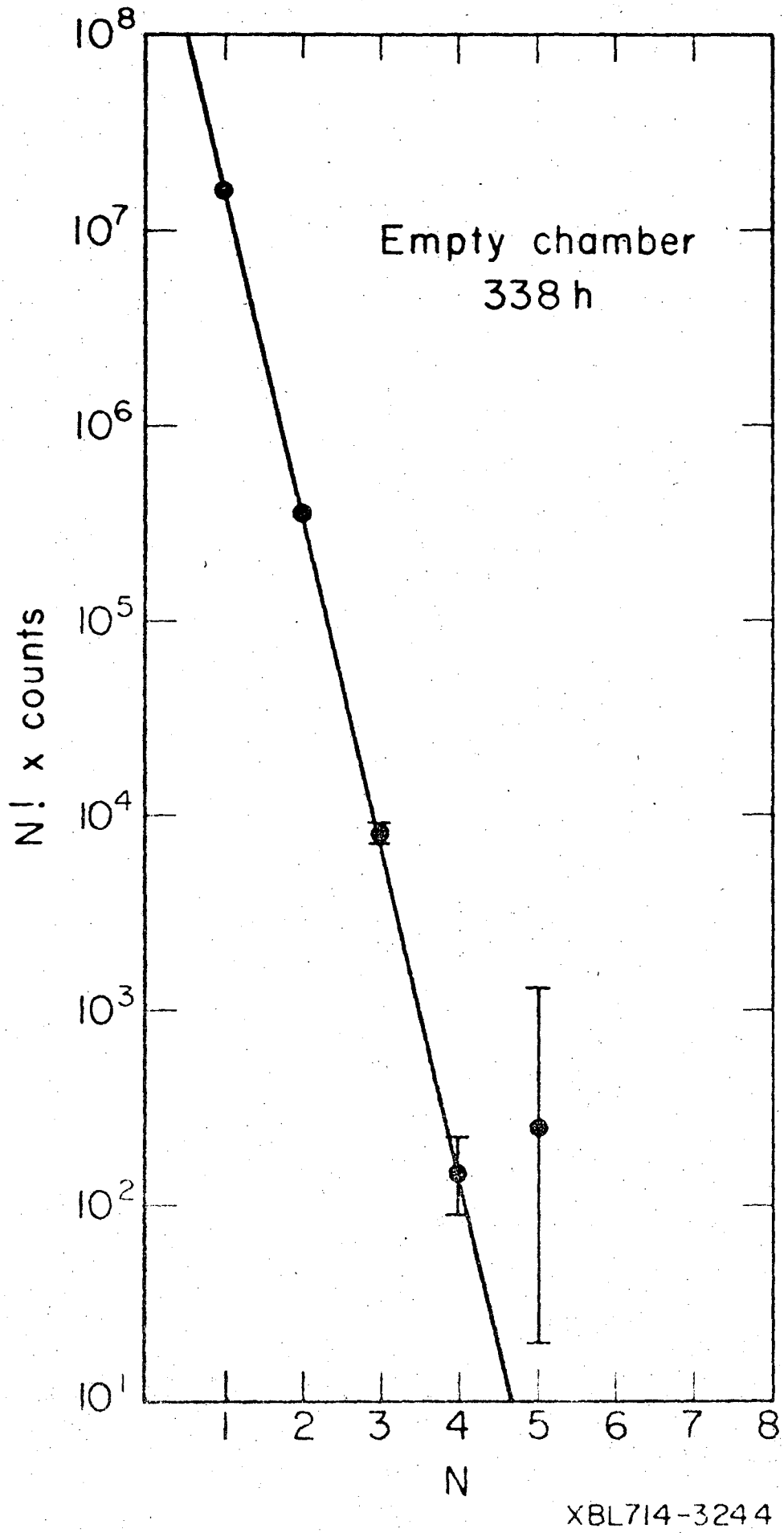

Empty chamber
338 h
N! x counts
N
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10^1
1
2
3
4
5
6
7
8
XBL714-3244

Fig. 5

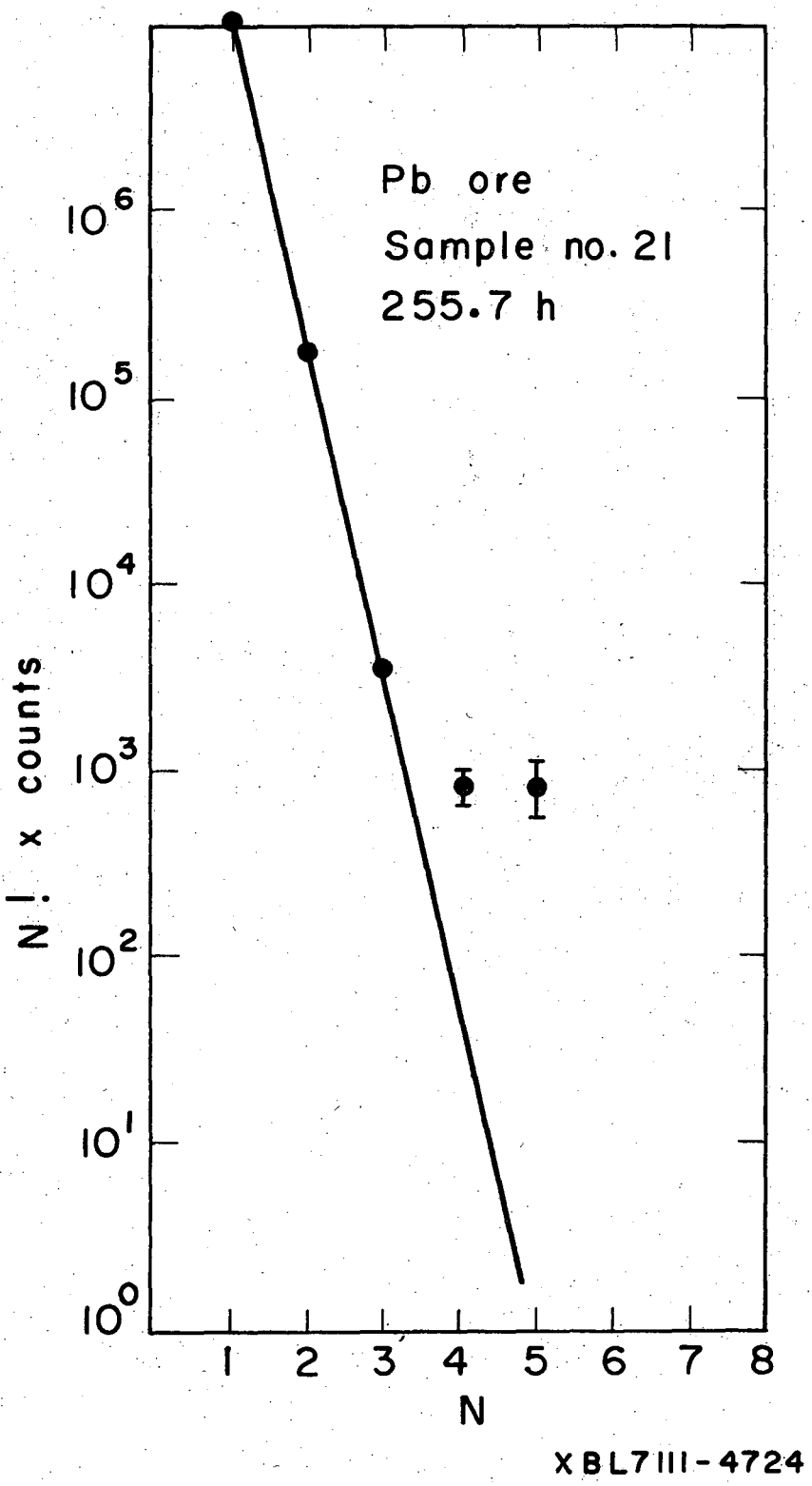

Pb ore
Sample no. 21
255.7 h
10^6
10^5
10^4
10^3
10^2
10^1
10^0
N! x counts
1
2
3
4
5
6
7
8
N
XBL7111-4724

Fig. 6

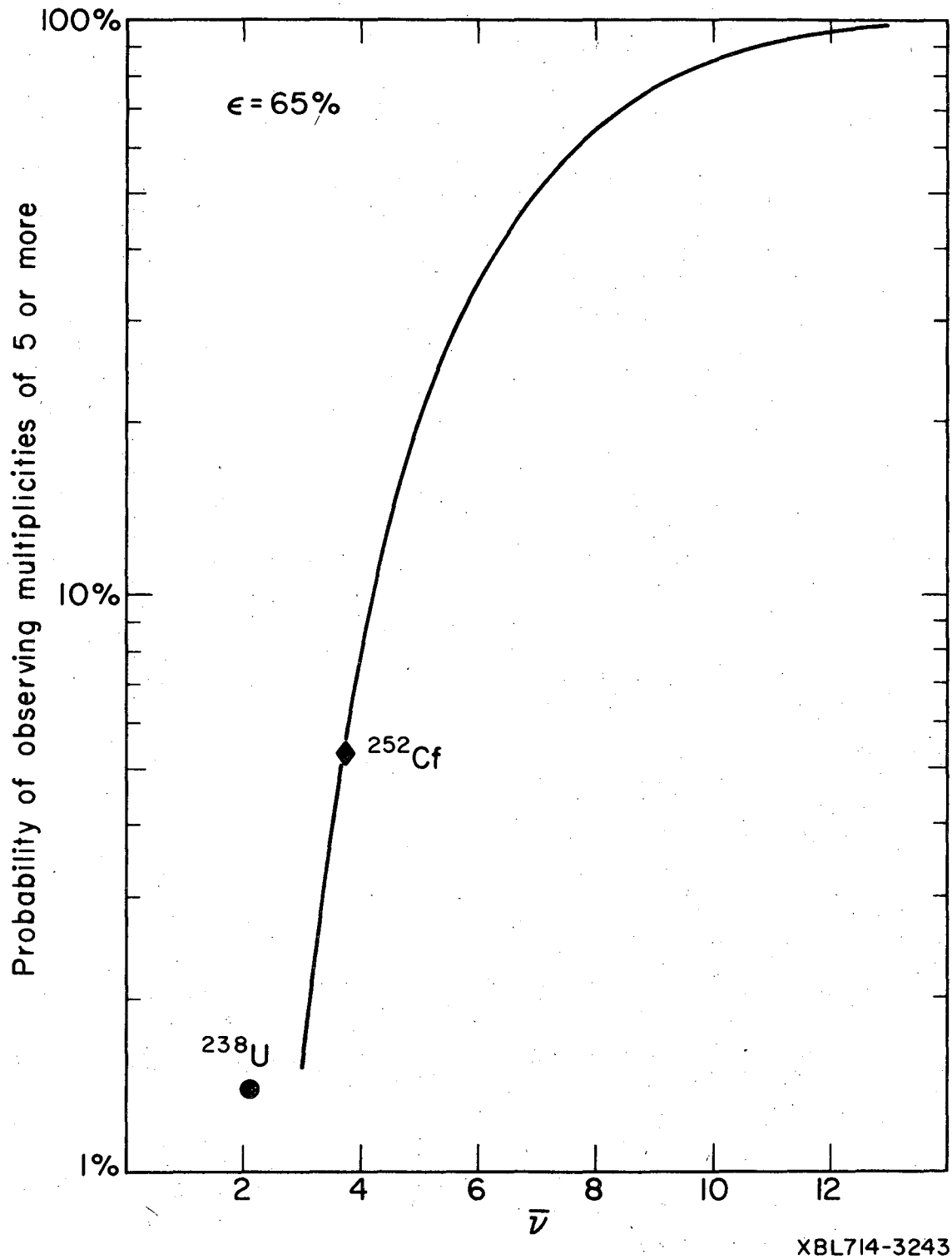

100%
10%
1%
ε = 65%
Probability of observing multiplicities of 5 or more
252Cf
238U
2
4
6
8
10
12
ν̄
XBL714-3243

Fig. 7

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LAWRENCE BERKELEY LABORATORY
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BERKELEY, CALIFORNIA 94720