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A COMPARISON OF METHODS FOR ASSAYING CALIFORNIUM-252

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A COMPARISON OF METHODS FOR ASSAYING CALIFORNIUM-252

by

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ABSTRACT

Three methods for assaying californium-252 are compared: neutron counting, alpha counting, and fission fragment counting. All three methods give consistent results; neutron counting is the most satisfactory method for routine use. Alpha counting requires correction for the counts contributed by fission fragments.

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INTRODUCTION

In producing californium-252 and subsequently processing and encapsulating this isotope for use as a neutron source, a variety of analytical methods are used to assay the californium. Encapsulated sources can be assayed by calorimetry, manganese bath activation, and neutron counting; for process solutions, the techniques of alpha counting or fission fragment measurement can also be used. These methods of analysis must be consistent with one another to assure process material balance and to enable the fabrication of sources of the required specific activity. The counting methods were compared to assure this consistency among the analytical methods of californium-252 radioassay used at Savannah River.

SUMMARY

Determinations of californium-252 by counting neutrons, alpha particles, and fission fragments agree within 1.3 percent of one another. Fission fragment measurements of a californium-252 sample made with a Frisch Grid Chamber agreed closely with those obtained using a "Lexan"* track recorder. An alpha to fission disintegration ratio of 31.3 was confirmed by measurements from a silicon detector and a Frisch Grid Chamber.

The alpha counters (gross and low geometry) detect both alpha particles and fission fragments. The counting efficiency for fission fragments is dependent on the density and chemical composition of solids on the plate.

Neutron counting is necessary for assaying larger californium-252 samples and is also the usual method for routine radioassay of small samples. Exceptions may arise in highly dilute solutions or in the assay of californium containing appreciable ^{254}Cf; in these cases, the use of the Frisch Grid Chamber is necessary to characterize the sample.

* Registered trademark of General Electric Co. for polycarbonate film.

DISCUSSION

Comparison Measurements

The californium solution used for comparison was obtained from californium recovery operations and was purified from other actinides (especially ^{244}Cm) before use. The entire sample was put into a vial, and its neutron emission rate was measured with the Mn bath neutron counter. (A summary of the counting equipment and counter calibrations used in this study is given in Appendices A and B.) Gross alpha counts of aliquots were made in triplicate on the LOGAC and PCC-10 counters. Alpha and fission fragment energy spectra of californium aliquots electrodeposited on nickel plates were obtained with the Si detector and the Frisch Grid Chamber. One such electroplated californium source was also used for fission determinations by the track recorder method. Aliquots of the solution were also counted on the neutron counter.

The relative response of neutrons and fission fragments were compared on the ZnS and PCC-10 detectors by counting the same plates on both counters.

The distortion of the alpha and fission fragment spectra by solids was studied by interposing a piece of mica, 2.3 mg/cm^2, between the sample and the Si detector. An electrodeposited source containing ^{233}U, ^{239}Pu, ^{238}Pu, ^{244}Cm, and ^{252}Cf was used for some of the alpha studies.

The "dead time correction" for the neutron counter was determined by counting aliquots of the californium solution. The curve in Figure 1 shows this correction, which is necessary because the counter is insensitive when a pulse is being processed.

Isotopic Abundance by Alpha Spectroscopy

Two different determinations were made of the relative alpha activities of the three californium isotopes in the purified californium solution, as follows: ^{252}Cf, 86.4% and 86.8% of total alpha activity; ^{250}Cf, 12.0% and 12.3%; and ^{249}Cf, 1.5% and 0.91%. (^{249}Cf is not resolved from any ^{244}Cm contamination remaining in the purified curium.) All calculations based on the alpha composition used 86.8% for ^{252}Cf. An alpha spectrum is shown in Figure 2.

Gross Alpha and Fission Fragment Counting

Counting californium-252 by alpha detectors requires corrections for the alpha contributions of other californium isotopes, and also for the detection of the two fission fragments produced during fission. Both the alpha particles and the fission fragments produce highly ionized paths in a counting medium, and counts will be registered for both types of particles in gross alpha counters. Spurious pulses can also be produced by the high energy pulses formed by the fission fragments. These pulses saturate the preamplifiers or amplifiers, and the pulse "overshoot" can be large enough to be counted as an additional alpha count. Any errors introduced in the gross alpha results would also be reflected in the neutron counter results because of the method of standardizing the neutron counter. Therefore, special attention was given to the fission fragment contributions to alpha counting in the various instruments used.

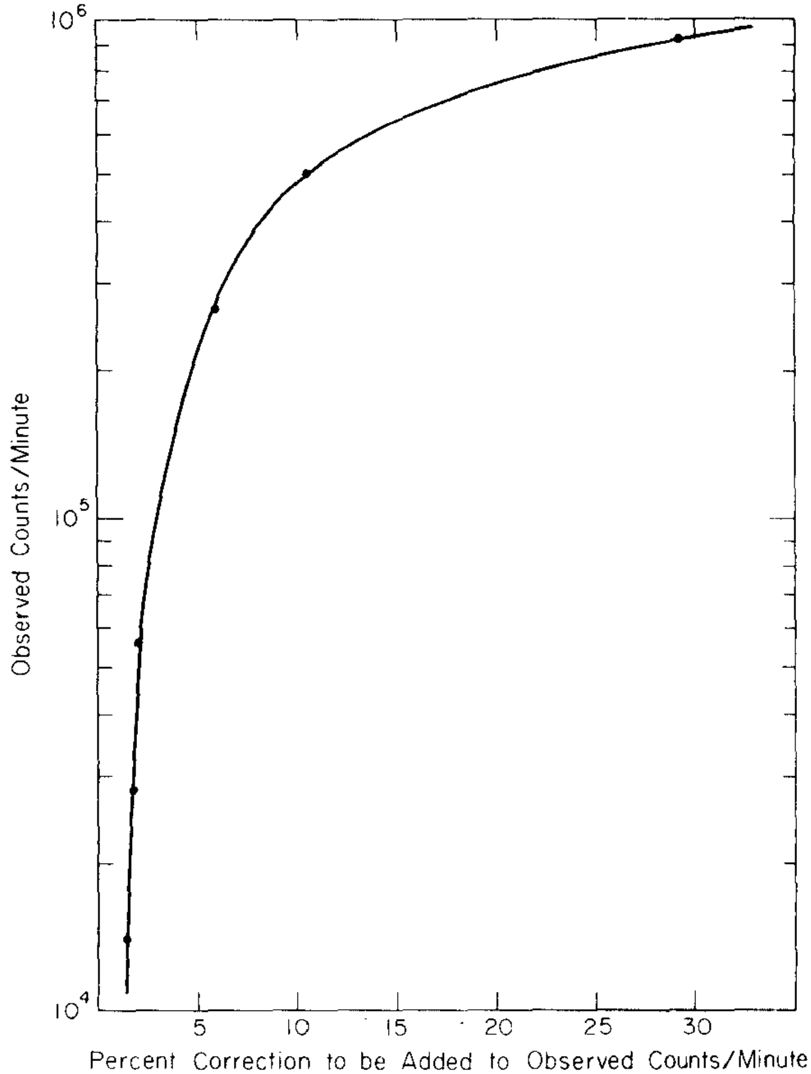

FIG. 1 CORRECTION FOR COINCIDENCE LOSSES IN NEUTRON DETECTOR

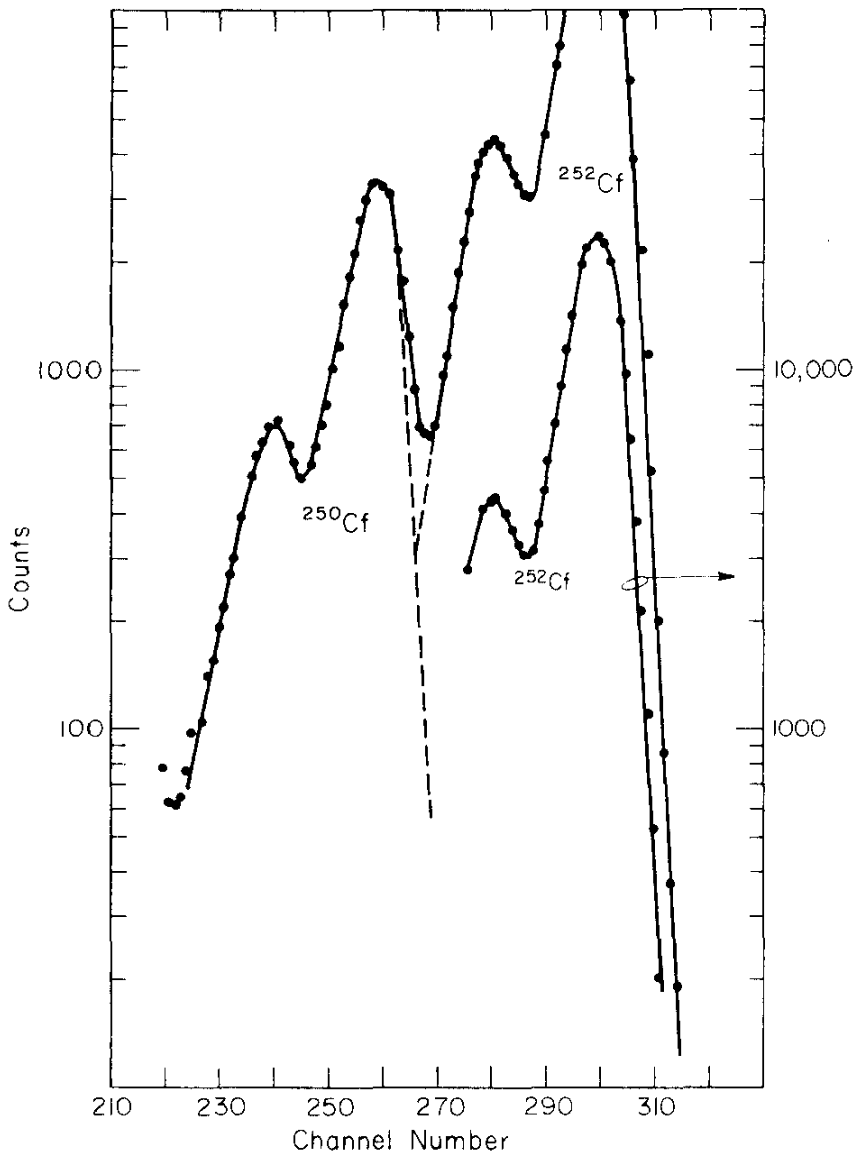

FIG. 2 ALPHA SPECTRUM OF PURE CALIFORNIUM

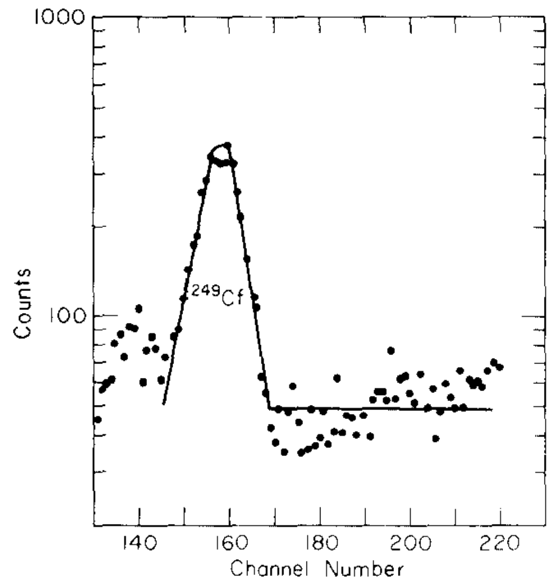

FIG. 2 (Continued)

The number of alpha particles and fission fragments being emitted per minute by the total californium solution was found to be $(1.796 \pm 0.004)10^9$ when counted with the PCC-10 counter, and $(1.776 \pm 0.006)10^9$ when counted with the LOGAC counter. The difference in the two values, 0.020×10^9, is more than twice the standard deviation of $\pm 0.007 \times 10^9$. The PCC-10 was calibrated against the LOGAC with ^{239}Pu sources, and no bias was observed with ^{244}Cm sources counted in both counters.

Counting in the LOGAC is done through a ~2.5 mg/cm^2 mica window, but there is no absorber in the PCC-10 counter. The lower count rate in the LOGAC might, therefore, be attributed to fission fragment absorption by the mica. In order to evaluate this absorption, the alpha and fission fragment spectra of a californium sample was observed by the Si detector with and without a mica absorber. Without mica, the fission fragment to alpha ratio was 0.052; with a mica absorber, the ratio was 0.039. These ratios would correspond to a difference ratio in total count (alpha + fission fragments) between the two counters of 1.052/1.039 = 1.012. This agrees closely with the observed difference ratio of 1.796/1.776 = 1.011.

The ZnS detector gives the same results as the PCC-10 for the californium alpha plus fission fragment count rate. This indicates that the ZnS detector counts fission fragments at about the same efficiency as the PCC-10.

Additional pulse height analysis studies were made to determine the effect of the mica absorber. The spectra of a mixed alpha source, as measured by the Si crystal with and without the mica absorber present are shown in Figure 3. The fission spectrum through mica was a continuum with counts in all channels from the maximum alpha energy, 6.12 Mev, up to an apparent alpha energy of about

14 Mev. The fission fragment spectrum without the mica (Figure 4) shows very few counts in the region from 6.12 to 9 Mev (apparent alpha energies), and the two peaks are centered at about 10 and 13 Mev. (The resolution of the high energy peak seems to be better because of the saturation of the preamplifier by the large pulses.)

A series of comparative alpha and fission fragment counts were made to correlate these results with the best available nuclear data (Appendix C). The purified californium was counted in the Frisch Grid Chamber and the Si detector to get a ratio of alpha particles to fission fragments. In a typical experiment, the Frisch Grid Chamber gave 599 fission fragments/min and 1.081×10^4 alpha particles/min for the electrodeposited californium source. The ^{252}Cf alpha particles, (0.868)(total californium alpha particles), give 9.393×10^3 counts/min. The counter geometry is very close to 100% for fission fragments and 50% for alpha particles, thus the alpha/fission ratio is $(1.877 \times 10^4)/599 = 31.3$. Four determinations were made with the Si detector, and an average value of 31.5 ± 0.65* was obtained. These values are close to 31.3 given in Appendix C.

In a second comparison, the electrodeposited source that had produced 599 fission fragments/min in the Frisch Grid Chamber was placed next to a piece of "Lexan," and the resultant fission tracks were counted. The value of 592 ± 18 tracks/min derived from a seven-minute exposure compared favorably with the result obtained with the Frisch Grid Chamber.

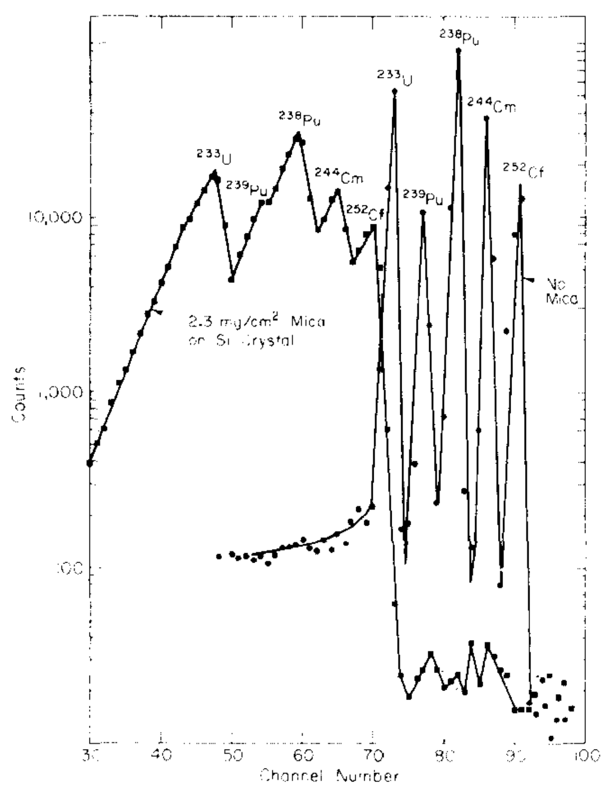

FIG. 3 ALPHA SPECTRA OF MIXED ALPHA SOURCE WITH AND WITHOUT MICA ABSORBER

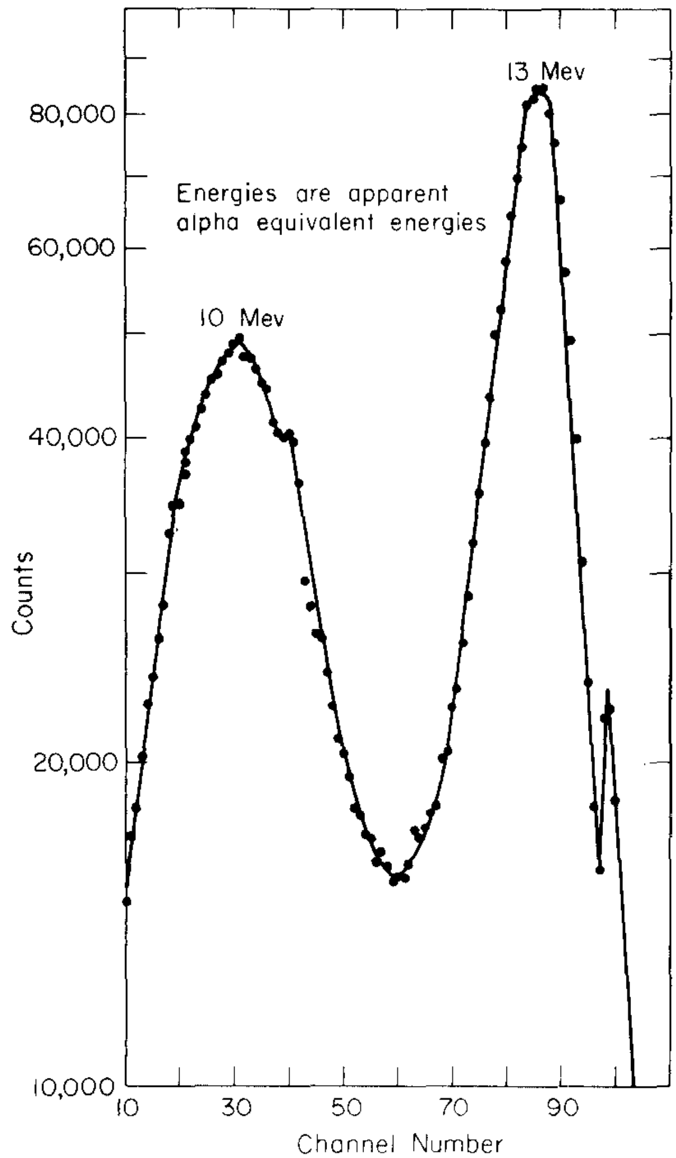

FIG. 4 SPECTRUM OF FISSION FRAGMENTS

Neutron Counting

The total solution of californium in the vial was assayed for ^{252}Cf by placing it in the Mn bath and counting the induced ^{56}Mn activity. A result of 1.786×10^8 neutrons/min was obtained.

An aliquot of the solution was then counted in the neutron counter; the result, when calculated back to the volume of the total solution, corresponded to 1.770×10^8 neutrons/min for the solution. This value agrees within 1% of that determined by the Mn bath.

Comparison of Neutron, Alpha, and Fission Counting

If the values of 31.3 alpha particles per fission and 3.8 neutrons per fission are used, the number of fissions and alpha particles can be calculated from the neutron emission rate and compared with those observed experimentally. With the neutron emission value of 1.770×10^8 for the total solution, the fission rate can be derived as $(1.770 \times 10^8) \div 3.8 = 4.658 \times 10^7$ min^{-1}, and the alpha rate is $(4.658 \times 10^7)(31.3) = 1.458 \times 10^9$ min^{-1}.

The observed alpha count of 1.796×10^9 min^{-1} obtained from the PCC-10 must be corrected for fission fragments and for the contribution of other californium isotopes. With the values previously determined (^{252}Cf alpha = 86.8%; alpha/fission ratio = 31.3 for this sample, from Frisch Grid Chamber counting), the corresponding amount of ^{252}Cf is calculated:

Total PCC-10 counts/min = ½ (Total alpha)

$$(\text{Total alpha counts/min})(0.868) = {}^{252}\text{Cf alpha}$$

$$\text{Total PCC-10 counts/min} = \frac{{}^{252}\text{Cf alpha}}{0.868} + \frac{({}^{252}\text{Cf alpha})(2.00)^{**}}{31.3}$$

$$^{252}\text{Cf alpha counts/min} = \frac{1.7958 \times 10^9}{1.216} = 1.477 \times 10^9$$

^{252}Cf alpha counts/min calculated from the neutron results, 1.458×10^9, differs from the above by 1.3%. The results calculated from the PCC-10 count rate are independent of the reported values of the alpha/fission ratio and the number of neutrons per fission, if the PCC-10 and the Frisch Grid Chamber are assumed to count the fission fragments and the alpha particles with the same relative efficiencies. The comparison therefore indicates that the two methods are consistent to within 1.3% when the values for nuclear constants (Appendix C) are accepted. The neutrons per fission are probably not known this well,[1] so that the methods are consistent for all practical purposes.

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* The error shown is the standard deviation from the mean for the four experiments.

** Value determined by absolute counting with standards calibrated on a low-geometry counter.

APPENDIX A

COUNTING EQUIPMENT

PCC-10:	A gross alpha counter of the gas flow proportional type using P-10 gas (90% argon – 10% methane), with 2π geometry.
ZnS Counter:	An alpha scintillation counter used as an alternate to the PCC-10 for routine counting. This detector has about 2π geometry.
LOGAC:	LOw Geometry Alpha Counter. A low geometry gas flow proportional counter used for absolute alpha standardization.[2] A 2.20 mg/cm^2 mica window separates the counting chamber from the evacuated source chamber.
Si Detector:	A gold surface-barrier silicon detector, 50 mm^2 area, resolution of 15 kev (FWHM).
Neutron Counter[3]:	A polyethylene-moderated well detector, with 10 BF_3 counting tubes. It has a 10% counting efficiency for ^{252}Cf neutrons.
Frisch Grid Chamber:	A P-10 gas-filled flow chamber designed for pulse height analysis. The grid suppresses the detection of positive ions, producing a peak proportional to the energy deposited in the gas. Resolution of 35 kev (FWHM) is obtained for alpha spectra.
Mn Bath[4]:	The Mn bath neutron counter consists of a 4-ft-dia by 4-ft-high tank filled with a saturated solution of manganous sulfate. The gammas emitted from ^{56}Mn (produced by $^{55}Mn(n,\gamma)^{56}Mn$) are counted in a NaI(Tl) crystal. The system has been calibrated with a Pu-Be source standardized by the National Bureau of Standards and also by a ^{252}Cf source cross-checked by the NBS.
Track Recorder[5]:	The track recorder method for fission fragment counting uses "Lexan" polycarbonate plastic sheet, which is placed in contact with the fissioning source, exposed to fission fragments, and then etched with NaOH. The resulting etched tracks are viewed with a microscope and counted visually.

APPENDIX B

COUNTER CALIBRATIONS AND CORRECTIONS

Calibrations for counters other than the Mn bath are referred ultimately to the LOGAC counter.[2] PCC-10 and ZnS gross alpha counters are calibrated directly against the LOGAC with ^{239}Pu sources. The neutron detector was calibrated with a known amount of ^{252}Cf, as determined by gross alpha counting. The fission track recorder method also requires a count of the ^{252}Cf on the source plate by one of the above counters.

APPENDIX C

NUCLEAR CONSTANTS OF CALIFORNIUM ISOTOPES

The physical constants for the decay of the three alpha-emitting isotopes present in the californium were obtained from References 6-8. Those used for this study are listed below.

Cf	*Total Half-life, years*	*Alpha Half-life, years*	*Alpha/fission*	*Alpha Energy, Mev*
249	360 [a]	360 [a]	4×10^6 [a]	5.812 (84%) [a]
250	13.2 [b]	13.2 [b]	1260 [b]	6.031 (83%),[a] 5.987 (17%)
252	2.621 [c]	2.73 [b]	31.3 [b]	6.119 (84%), [a] 6.076 (16%)

a. Reference 6
b. Reference 7
c. Reference 8

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