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FISSION-PRODUCT YIELDS FOR THERMAL-NEUTRON FISSION OF ^{243}Cm DETERMINED FROM MEASUREMENTS WITH A HIGH-RESOLUTION LOW-ENERGY GERMANIUM GAMMA-RAY DETECTOR

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ABSTRACT

Cumulative fission-product yields have been determined for 13 gamma rays emitted during the decay of 12 fission products created by thermal-neutron fission of ^{243}Cm. A high-resolution low-energy germanium detector was used to measure the pulse-height spectra of gamma rays emitted from a 77-nanogram sample of ^{243}Cm after the sample had been irradiated by thermal neutrons. Analysis of the data resulted in the identification and matching of gamma-ray energies and half-lives to individual radioisotopes. From these results, 12 cumulative fission product yields were deduced for radionuclides with half-lives between 4.2 min and 84.2 min.

INTRODUCTION

Data on cumulative fission-product yields have been published for fissioning systems ranging from ^{228}Th to ^{258}Fm, though studies thus far have concentrated primarily on the U, Pu, Cm, and Cf systems.[1] In the case of thermal-neutron fission, the mass distribution of fission products is asymmetric in all but the heaviest isotopes. This mass asymmetry appears as two peaks separated by a minimum occurring at a mass value corresponding to a symmetric division of the fissioning nucleus. The principal change in the distribution with heavier systems is a shift in the light-mass peak toward heavier mass values. The heavy-mass peak remains relatively fixed, though both peaks exhibit some broadening.[1] A consistent and accurate theoretical description of this behavior does not exist presently, though there are some semi-empirical models.[2] Hence there is motivation for acquiring further data, particularly for systems not previously investigated, to provide corroboration with theoretical models as they are formulated.

Toward this goal, J. K. Dickens and J. W. McConnell conducted a series of experiments in 1980 at Oak Ridge National Laboratory (ORNL) to determine the fission-product yields resulting from thermal-neutron fission in a sample of ^{243}Cm. Several sets of data were accumulated, the first of which has been analyzed and reported by D. G. Breederland.[3] The present report details the evaluation of a second data set which, though there is a slight overlap, has rather different characteristics from the first.

EXPERIMENTAL DETAILS

In July 1980 a 77-nanogram sample of curium nitrate, enriched to >99% in ^{243}Cm, was irradiated with thermal energy neutrons from the High Flux Isotope Reactor at ORNL. The data accumulated following this irradiation included those reported by Breederland.[3]

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In December 1980, after a cooling period that allowed the shorter-lived fission products to decay below significant levels, the same sample was irradiated again at the Oak Ridge Research Reactor, this time for 180 s in a 5×10^{13} n/cm^2/s thermal-neutron beam (thermal/resonance larger than 30) producing 1.06×10^9 ($\pm 7.5\%$) fissions (in the sample). Fifteen gamma-ray spectra were collected using a high-resolution intrinsic germanium detector configured to study the low-energy portion of the gamma-ray spectrum. The source-to-detector distance was 3 cm. One of these spectra is presented in Figs. 1a and 1b. The two peaks in Fig. 2 illustrate the high-resolution aspect of this detector, both peaks remaining well-defined with a resolution (full-width at half-maximum) of 0.55 keV at E_γ = 190 keV. The measurements began about 11 min after the end of irradiation and were terminated 21 hours later.

DATA REDUCTION AND ANALYSIS

The gamma rays emanating from the sample subsequent to fission result from decay transitions in the fission products which were produced either during irradiation or later by decay from other fission products. Identification of the individual unseparated products was accomplished by comparing peaks in the gamma-ray spectra with known decay transition energies for various radioisotopes.

Much of the work of reducing the spectral data was accomplished with two computer codes, TPASS[4] and ANGAM[5]. TPASS locates peaks in the spectrum, determines their areas, corrects for detector efficiencies, and attempts to identify sources of the peaks by comparisons with a table of radionuclide decay data. In the present analysis, energies were matched to within 0.5 keV. Peak identifications assigned by TPASS are exhibited in Figs. 1a and 1b.

In locating peaks, the code assumes a simple analytic shape and performs a parametric search on only the Gaussian portion of the peak. After finding as many peaks as possible on the first pass through the spectrum, the code parameterizes the resolution width as a function of E_γ. The intrinsic resolution widths, $\sigma(E_\gamma)$, for the detector used are known for ideal conditions. TPASS allows for minor variations in $\sigma(E_\gamma)$ resulting from non-ideal counting conditions, and then flags peaks with widths apparently too large to be attributed to a single gamma ray. As a result of fixing $\sigma(E_\gamma)$, unresolved doublets and some partially-resolved multiplets are adequately analyzed.

Uncertainties associated with the peak areas are derived from the estimated minimum continuum subtraction rather than "statistically." The reason for this approach is that the continuum is not a random distribution, but rather arises from two phenomena: Compton scattering of high-energy gamma rays in the detector itself; and the effects of data being spread over several channels by the finite resolution of the data-taking equipment. The algorithm used to calculate this "background" continuum tends to slightly overestimate the subtraction, and thus it is occasionally necessary to adjust the background values to obtain consistent results. The present data reduction followed very closely the methods detailed by Breederland.[3]

After the gamma-ray peaks were ascribed to their various isotopes, the identification was further validated using the program ANGAM to analyze the time-dependency of the yields of individual peaks. ANGAM deduces the apparent decay constant of a given gamma-ray yield and compares it with that of the assigned fission product. For some isotopes it was necessary to include a growth factor to account for the decay of parent isotopes. An example is illustrated by Fig. 3, which shows the decay of ^{131}Te into ^{131}I. The ^{131}Te isotope could be produced directly during fission; it could also be produced by decay of its parent, ^{131}Sb, if its parent is also produced during fission. The dashed line represents a case in which only ^{131}Te and no ^{131}Sb is initially present in the sample. The solid line represents a "best

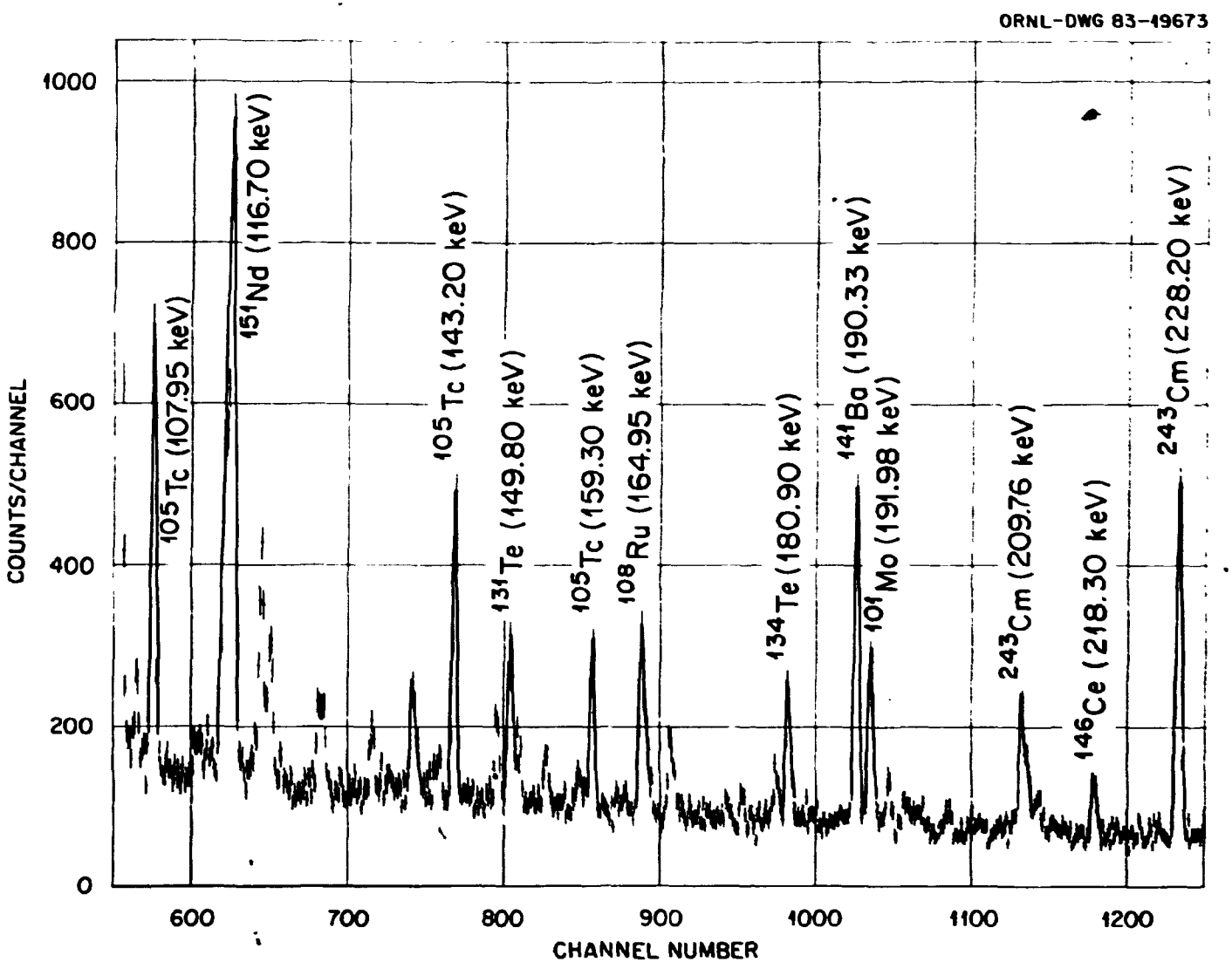

Fig. 1a. Lower-energy portion of the gamma-ray spectrum emitted following a 180-s thermal-neutron irradiation of a ^{243}Cm sample. The portion of the spectrum below 100 keV contained mostly x-ray peaks. For this spectrum, the cooling time was 670.2 s and the counting time was 200 s. The source-to-detector distance was 3 cm.

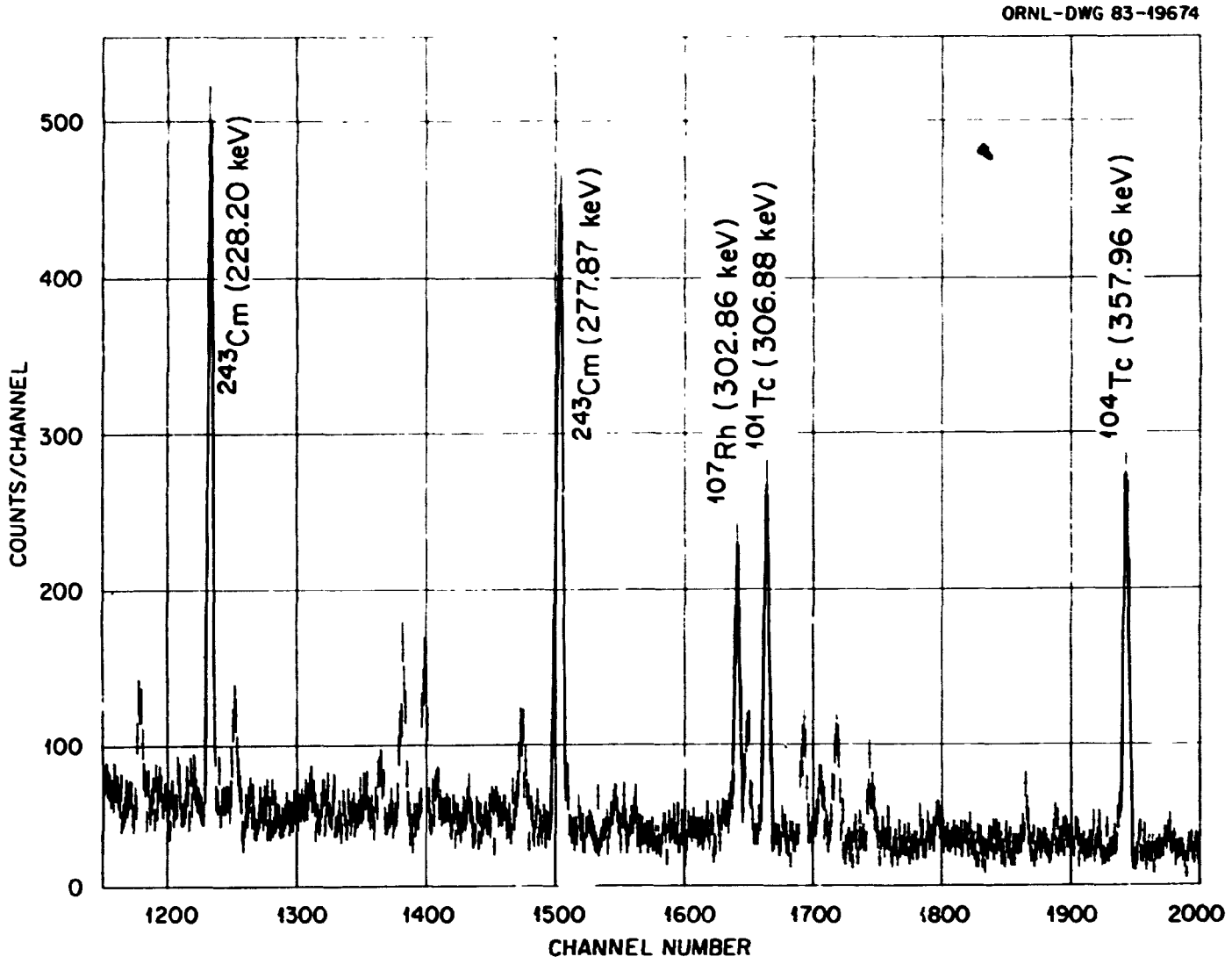

Fig. 1b. Higher-energy portion of the gamma-ray spectrum of Fig. 1a.

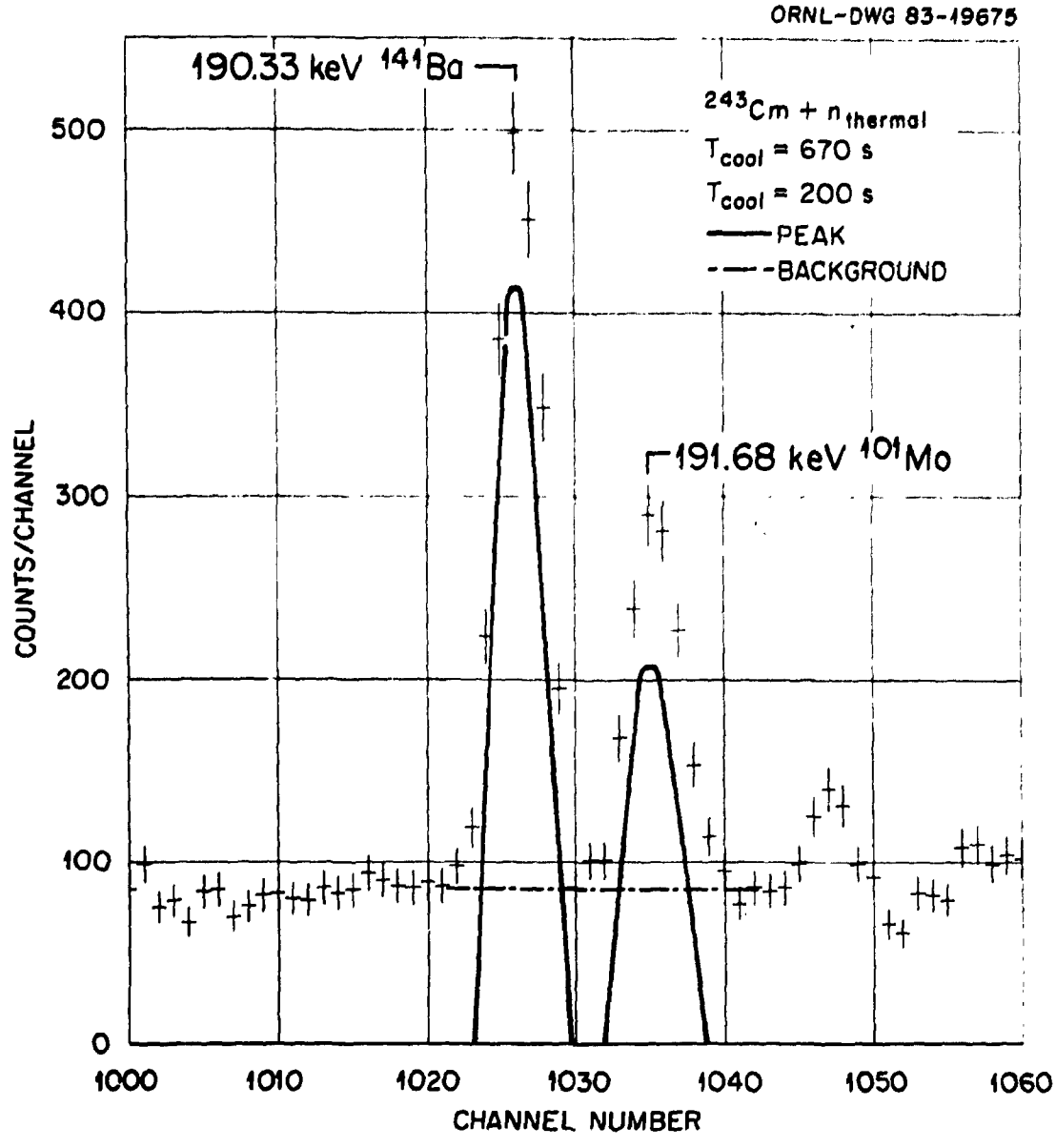

Fig. 2. Portion of a pulse-height spectrum of gamma rays illustrating the resolution capabilities of the germanium detector used. The solid lines represent the gamma-ray peaks after background continuum subtraction.

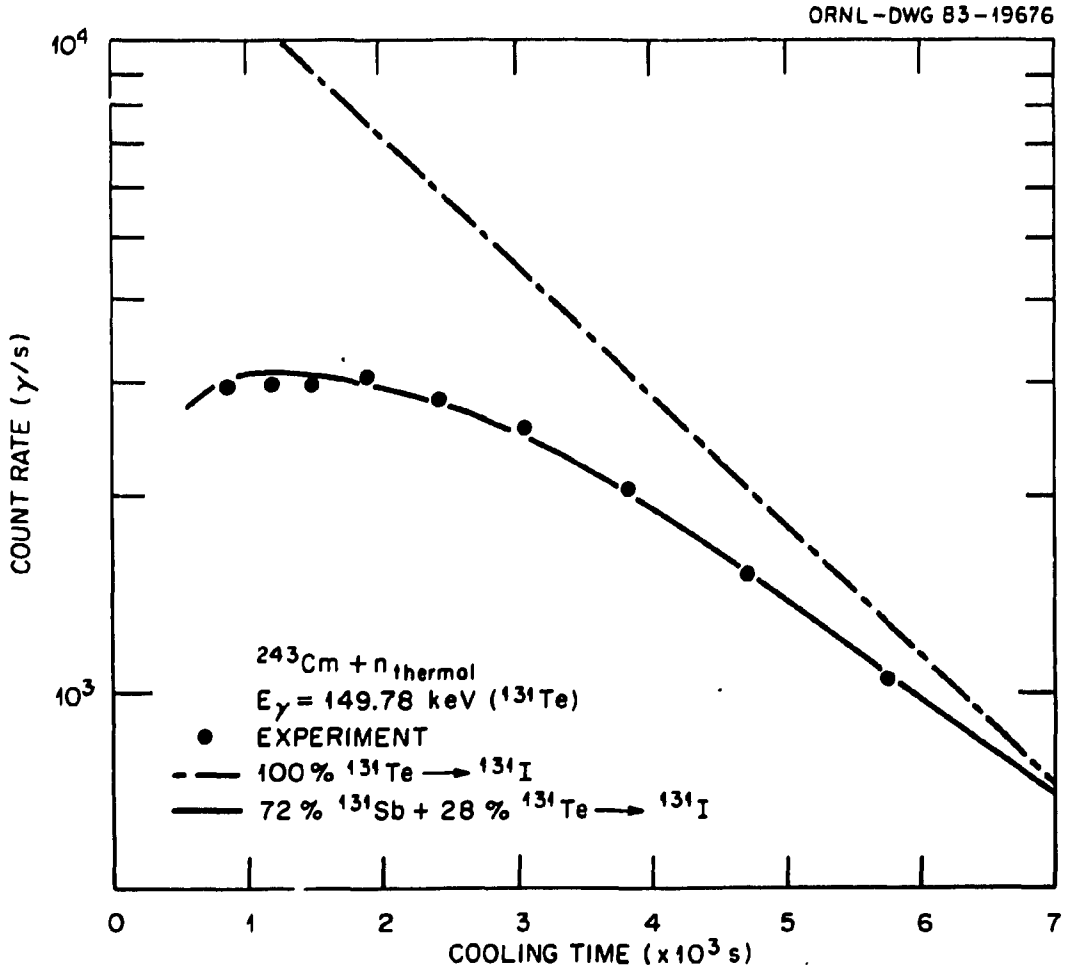

Fig. 3. Decay curves showing the growth effects due to the presence of both ^{131}Sb ($T_{1/2}$ = 23 min) and its daughter, ^{131}Te ($T_{1/2}$ = 25 min), immediately after fission. As the amount of ^{131}Sb dwindles, the combined curve asymptotically approaches the decay curve expected if only ^{131}Te were produced during fission.

fit" to the data observed for the case, in which both ^{131}Te and its parent ^{131}Sb were present immediately after fission. The curve shows the additive effects produced by the growth of ^{131}Te due to the decay of ^{131}Sb.

To calculate cumulative fission-product yields from the estimated gamma-ray yields, it is necessary to include gamma-ray branching ratios,[1,6,7] detector efficiency, and the half-lives of both the radioisotope being considered and its parent isotope, if there is one. A least-squares algorithm is used to fit the time-dependent gamma-ray yields, Y(t), to a decay curve and to project the effective yield immediately after fission. The fission-product yield was then obtained by dividing this effective yield by the gamma-ray branching ratio and by the number of fissions. This final value represents the fraction of the total fissions which produced a particular fission product.

Uncertainties in the cumulative fission-product yields were obtained by quadratically combining the uncertainties in (1) the gamma-ray yields, (2) the number of fissions, and (3) the gamma-ray branching ratios. The uncertainties in gamma-ray yields varied with the counting statistics for each gamma ray, but were typically less than 2%. The uncertainty in the number of fissions was evaluated to be 7.5%. The uncertainties assigned to the gamma-ray branching ratios vary widely and are included in Table 1, along with the other data discussed here.

Table 1. Gamma-ray intensities associated with decay of fission products following thermal-neutron fission of ^{243}Cm and the deduced cumulative fission-product yields.

E_γ (keV)	Yield per 100 fissions	Assigned fission product	Gamma-ray branching ratio (%)[a]	Cumulative fission-product yield (%)
107.69	0.65 ± 0.05	^{105}Tc	9.9 ± 1.3[b]	6.59 ± 1.0
140.51	4.71 ± 0.36	^{99}Tc*	87.2 ± 0.7	5.41 ± 0.42
143.26	0.84 ± 0.06	^{105}Tc	11 ± 1	7.64 ± 0.88
149.79	1.44 ± 0.11	^{131}Te	68 ± 1	2.10 ± 0.16
165.21	0.89 ± 0.08	^{108}Ru	18 ± 7	3.19 ± 0.85
165.94	1.47 ± 0.39	^{139}Ba	23.8 ± 0.3	6.22 ± 1.79
190.44	2.33 ± 0.18	^{141}Ba	46 ± 3	5.06 ± 0.51
192.12	1.03 ± 0.08	^{101}Mo	19.6 ± 4[b]	5.49 ± 1.20
218.34	0.50 ± 0.04	^{146}Ce	20.5 ± 3.2	2.45 ± 0.43
249.93	5.52 ± 0.42	^{135}Xe	90 ± 3[c]	6.14 ± 0.51
258.64	0.93 ± 0.07	^{138}Xe	31.5 ± 1.3	2.94 ± 0.25
303.00	3.64 ± 0.28	^{107}Rh	65.9 ± 4.6	5.52 ± 0.57
358.25	6.09 ± 0.46	^{106}Tc	89 ± 5[c]	6.84 ± 0.64

[a]Nuclear data taken from ref. 1, except as noted.
[b]Values from ref. 7.
[c]Values from ref. 6.

EVALUATION

Because of the relatively short counting intervals of the present data set, the fission products identified were primarily those which had half-lives between 4.2 min and 84.2 min and which decayed with gamma-ray energies ranging from 107.69 to 358.25 keV. (These contrast with the data reported by Breederland, which consisted of isotopes with half-lives from 6 hours to 65 days and gamma-ray energies from 140 to 1600 keV.[3]) Except for ^{105}Tc, only one gamma ray, usually the one with the largest branching ratio, was analyzed for each fission product. Yields were not obtained for all the nuclides within the range studied, some of which decay mostly by beta-ray emission. Others decay with a primary gamma ray with an energy very close to a more intense gamma ray from another fission product with a similar half-life. Such was the case for the 255.60-keV peak of ^{151}Nd and the 255.59-keV peak of ^{142}Ba. Several gamma rays had an E_γ indistinguishably close to the very strong ^{243}Cm peaks at 99.53, 103.73, 209.76, 228.20, and 277.87 keV, for example, gamma rays emitted during decay of ^{134}Te (E_γ = 210.57 keV, 277.72 keV) and ^{152}Nd (E_γ = 278.33 keV).

CONCLUSIONS AND COMPARISONS

In all, 23 gamma-ray peaks were analyzed and from these a total of 12 isotopes were identified sufficiently to allow adequate deduction of their cumulative yields. The results obtained for these isotopes are listed in Table 1 and are displayed graphically in Figs. 4a and 4b. The graphs include data points (inverted solid triangles) for those fission products evaluated earlier by Breederland, and the two sets appear to be generally in good agreement, with the possible exception of the value for ^{108}Ru.

In addition, two nuclides appearing in Table 1, but not in Fig. 4b, bear further comment. The cumulative yield of 2.10% reported for ^{131}Te is clearly less than that which would be in "agreement" with the mass-distribution curve. However, ^{131}Te has a comparatively long-lived isomeric state, decaying with a primary gamma-ray energy of 852.2 keV, and having a reported yield of 1.25% (±0.06%).[3] Both Te states decay to ^{131}I and the sum of their yields is in reasonable agreement with that reported for ^{131}I.[3] The other questionable value is the 2.94% yield of ^{138}Xe. The information extracted during the analysis of this isotope seems adequate to deduce an accurate yield value. Thus, the value is being reported, though no explanation is presented at this time for the apparently low yield.

Also plotted in Figs. 4a and 4b are values for the mass distribution of ^{245}Cm from an earlier ORNL study.[1] The shifting of the light-mass peak toward heavier mass values is evident in the ^{245}Cm distribution. The heavy-mass peak does indeed seem fixed, as predicted from the empirical systematics discussed in the Introduction. Possible broadening of the heavy-mass peak is not as apparent, perhaps because of the lack of ^{243}Cm data in the 110 to 130 mass value range.

For some of the data points shown in Figs. 4a and 4b the main source of uncertainty is in the raw data, the uncertainties arising from situations such as small peaks on a large "background" continuum. In other cases the greatest source of error lies with the nuclear data, primarily with gamma-ray branching ratios, some of which have rather large uncertainties.

Together with the Breederland[3] report, these are the only published data for ^{243}Cm fission-product yields, so a comparison with other studies of the same type is not possible. However, the portion of the data thus far analyzed does seem to reinforce the empirical structure which is seen in other systems.

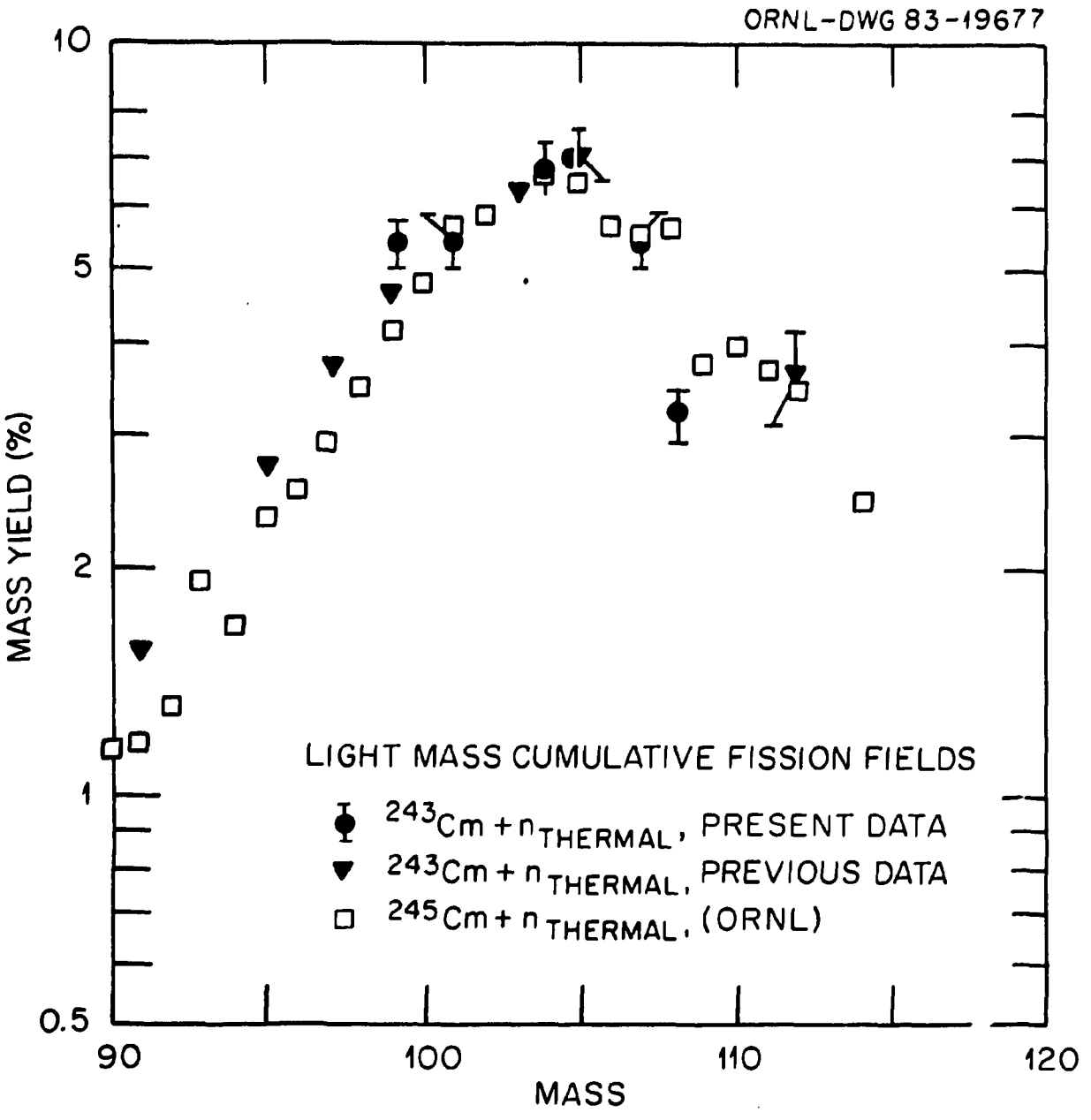

Fig. 4a. Light-mass fission-product yield distribution for $^{243}Cm + n_{thermal}$ deduced from present data. Shown for comparison are those data analyzed earlier as part of this same project (ref. 3), as well as data obtained from measurements of $^{245}Cm + n_{thermal}$ fission-product yields (ref. 1). The ^{245}Cm data are shifted toward heavier mass values.

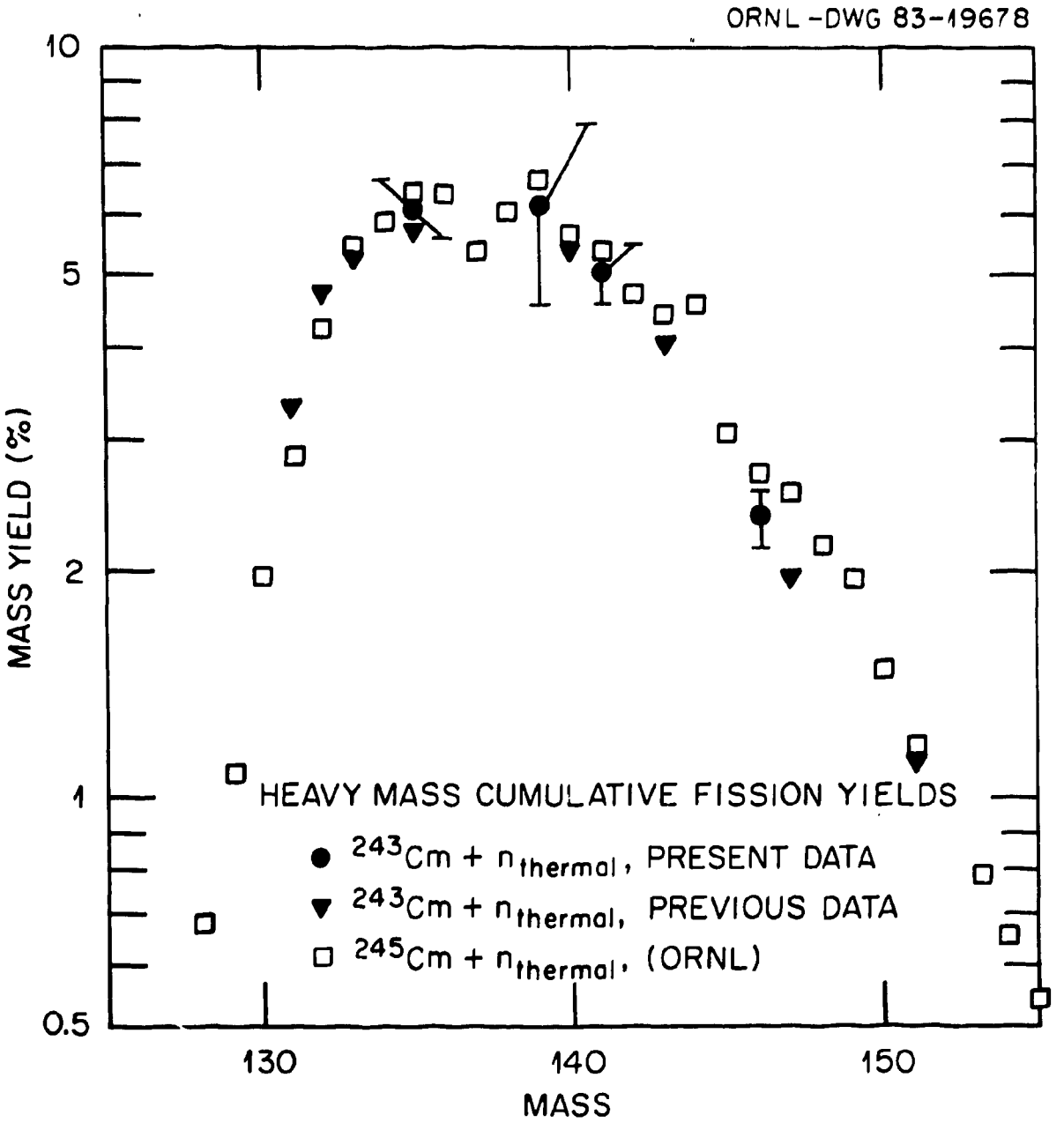

Fig. 4b. Heavy-mass yield distribution for $^{243}Cm + n_{thermal}$ deduced from present data. The mass distribution is similar to that for $^{245}Cm + n_{thermal}$, which is shown for comparison.

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