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ISOTOPIC CONSTITUTION OF PLUTONIUM: PART III

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Abstract

This is the third and terminal report on the isotopic composition of plutonium. Data are reported on three samples of plutonium irradiated at Hanford. The last and most highly irradiated of these contained almost 7% Pu^{240}. The existence of the isotope Pu^{241} is established and its abundance measured. Evidence is presented for the existence of the isotope 95^{241}. A final table of data obtained with the mass spectrometer and the correlation with the spontaneous fission count on these samples is presented.

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Second Examination of CW-1B

In the last report of this series (LA-327), data on a sample of plutonium irradiated at Hanford, designated as CW-1B, were reported. The value obtained at that time (March 13, 1945) for the abundance ratio Pu^{240}/Pu^{239} was $(6.51 \pm 0.20) \times 10^{-3}$. The sample was reported to have been contaminated with $PbCl_2$. This was established by identification of ions of the types Pb^+, $PbCl^+$, $PbCl_2^+$. The chloride ions rapidly disappeared, presumably due to reduction of $PbCl_2$ to Pb by molten plutonium. However, an extraneous peak remained at mass position 241. The ratio of this peak height to that at 239 increased rapidly with increase of temperature of the microfurnace from which the sample was being distilled. The 241 peak persisted throughout the run until the plutonium sample was exhausted.

Since that report, a second examination of CW-1B has been made (November 12, 1945). The furnace with the sample in it was outgassed for about an hour at 1000° C before the first run was made. No peaks due to ions of the form of $PbCl^+$ or $PbCl_2^+$ were observed, but the lead spectrum (due to Pb^+) was found. A small peak was observed at mass position 241. The sample was allowed to stand in the tube (continuously evacuated) for about two weeks while instrumental difficulties were solved. On reexamination, a number of small extraneous peaks of unknown origin appeared. These peaks had not been observed in the first examination. The heights of these peaks decreased rapidly with time of distillation and disappeared. The abundance ratio Pu^{240}/Pu^{239} was then determined and a value of $(6.50 \pm 0.10) \times 10^{-3}$ obtained (cf above). Determination of the [redacted]

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Pu^{241}/Pu^{239} yielded the value $(8.1 \pm 0.5) \times 10^{-5}$. This quantity showed a slight tendency to decrease with distillation as though all the superimposed peak, one of those mentioned above, had not "distilled away." Thus, this latter value must be regarded as an upper limit only.

The behavior of the peak at mass position 241 was entirely different during the second examination than during the first. No complete explanation of the appearance and persistence (in the first examination) of a large peak at mass 241 has been found and this behavior has never been observed in any other sample.

Possible compound ions which could have been responsible for this behavior were discussed in LA-327. Only one of these ions remains as a possible cause of the 241 peak in the first examination of CW-1B. Since the phenomenon did not repeat itself on the second examination, actual contamination of the sample by extraneous compounds or metals is ruled out. During the first examination of CW-1B, the pressure in the spectometer tube was about ten times higher than is normally the case (1×10^{-6} mm of Hg instead of 1×10^{-7} mm). It was subsequently found that there was a pinhole in the plate lead of the ion gauge which caused this increase in pressure. Thus, there exists the possibility that the behavior of the 241 peak in this case could have been caused by a vapor-phase reaction of water vapor and plutonium vapor to form PuH_2 which might have a long enough lifetime to be ionized and travel up the spectrometer tube to the collector. This time is, of course, very short. It requires about 25 microseconds for a 2000 volt singly charged plutonium ion to

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travel one meter. No attempts have been made to reproduce these experimental conditions to see if this 241 peak behavior can be duplicated.

Sample CW-2

Sample CW-2 is the second of the samples irradiated at Hanford. This was a sample of Clinton plutonium which was placed in a Hanford pile on September 27, 1944, and was removed on April 12, 1945. It had received an irradiation, nvt, of approximately 8×10^{19}. This sample was decontaminated and reduced to metal by Seaborg at Chicago, then shipped to Los Alamos for mass spectrometric examination.

The first run across the 235-245 mass region, made immediately after turning on the microfurnace, showed a peak at mass position 241 which was only slightly smaller than that at 240. After a set of Pu^{240}/Pu^{239} abundance ratios had been determined, another curve of this region showed that the peak at 241 was less than 10% as high as that at 240. Twenty determinations of the abundance ratio Pu^{241}/Pu^{239} were made. The temperature of the microfurnace was varied for each set of five determinations. This ratio stayed constant within the limits of error of the measurement. The value $(2.14 \pm 0.05) \times 10^{-3}$ was obtained. The abundance ratio Pu^{240}/Pu^{239}, mentioned above, gave a value of $(4.64 \pm 0.05) \times 10^{-2}$. These determinations were made on August 25, 1945.

Seven days later, on September 1, 1945, twelve more sets of the ratio Pu^{241}/Pu^{239} were run, the spectrometer tube having remained evacuated for this period. The result was a value of $(2.09 \pm 0.06) \times 10^{-3}$.

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Peaks were observed at mass positions 255, 256, and 257. The ratio of the height of the peak at 257 to that at 255 was determined to be $(4.10 \pm 0.08) \times 10^{-3}$.

The above observed facts are interpreted in the following manner. The "large" 241 peak which rapidly decreased in height with time of distillation of the sample was produced by 95^{241+}. This material had grown from Pu^{241} (see below) by β decay. The rapid decrease in intensity with time shows that the vapor pressure of this material is much larger than that of plutonium. This is confirmed by an experiment by Seaborg at Chicago (CS-3312) in which a mixture of 95^{241} and plutonium was distilled in vacuo and condensed on a series of plates at different temperatures. Analysis of the condensates by means of the different range of the alpha particles from the two elements led to the conclusion that 95 has a vapor pressure of about 100 times the vapor pressure of plutonium at 1100°C.

The part of the 241 peak which remained after the 95^{241} has distilled away was produced by the plutonium isotope 241. This is reasonably certain, since the ratio of this peak height to that at 239 remained constant both with time and temperature of distillation of the sample.

The measurement of the ratio Pu^{241}/Pu^{239} a week after the first determination shows that the half-life of Pu^{241} is greater than 4 - 6 months.

The peaks at mass position 255 and 256 were caused by ions $Pu^{239}O^{16+}$ and $Pu^{240}O^{16+}$. No accurate determinations of the ratio of the height of these two peaks was made. A rough determination gave nearly the

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correct value. The peak at 257 was produced by the superposition of the ions $Pu^{239}O^{18+}$ and $Pu^{241}O^{16+}$. Smythe has determined* the abundance ratio O^{18}/O^{16} to be $(1.99 \pm 0.04 \times 10^{-2})$. Thus, if the contribution of the $Pu^{239}O^{18+}$ ion is substracted from the value 0.00410 recorded above, the value 0.00211 is obtained for the ratio $Pu^{241}O^{16}/Pu^{239}O^{16}$, confirming the direct determination of the ratio Pu^{241}/Pu^{239}. This gives strong support both to the interpretation that the 241 peak was caused by a plutonium isotope 241 and to the above interpretation of the cause of the 257 peak.

On February 19, 1945, a second sample of CW-2 was examined. This was 178 days after the first determination of the Pu^{241}/Pu^{240} abundance ratio on the first sample of CW-2.

On the first run across the plutonium peaks, the peak at 241 was about one-third as high at that at 240. On the second run, made immediately after the first, approximately 15 minutes of distillation having taken place, the above ratio had dropped to about 1/15. On the third run, immediately following the second, this ratio measured roughly 1/17. Not more than a total of 45 minutes were required for these runs.

Approximately one hour was required to make 10 determinations of the Pu^{240}/Pu^{239} abundance ratio. A value of $(4.66 \pm 0.05) \times 10^{-2}$ was obtained.

Approximately two hours were required to make 19 determinations of the Pu^{241}/Pu^{239} abundance ratio. This ratio stayed constant within the limits of error during this time. The data were taken at three temperatures of distillation of the sample and a value of $(2.06 \pm 0.08) \times 10^{-3}$ was obtained. About one-third of the sample was distilled away during these

* Phys. Rev. 45, 299 (1934)

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experiments.

These results confirmed those obtained five months before and increases the minimum value of the half-life of Pu^{241} to at least 4 years. This value was calculated by adding the errors to the abundance ratios in such a way as to give the greatest possible difference between these ratios (obtained 178 days apart).

A curve showing peaks at mass positions 241 and 257 is shown in Fig. 1. This curve was drawn from data obtained frpm Sample CW-2.

Sample CW-3

Sample CW-3 is the third of the plutonium samples which was irradiated at Hanford. This was a sample of Clinton plutonium which was placed in a Hanford pile on September 27, 1944, and was removed on June 14, 1945. It has received an irradiation, nvt, of approximately 1.3×10^{20}. This sample was decontaminated and reduced to metal by Seaborg at Chicago, then shipped to Los Alamos for mass spectrometric examination.

During the first two runs over the plutonium peaks, the large peak at mass position 241, which had been observed in Sample CW-2 to decrease rapidly in height, was not noticed. This was probably due to the fact that a ralatively short time had elapsed between the time the sample was purified chemically and the time of the mass spectrometric examination. Thus, relatively little Pu^{241} had decayed into 95^{241}.

Fifteen determinations of the abundance ratio Pu^{240}/Pu^{239} yielded the value $(7.39 \pm 0.08) \times 10^{-2}$.

Curves of the plutonium region run at much higher intensities showed two new peaks at mass positions 240 1/2 and 241 1/2, the former

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about ten times as intense as the latter. The ratio of these peak heights to one of the plutonium isotope peak heights was not constant, but increased rapidly with the temperature of the microfurnace. At the distillation temperature at which the Pu^{240}/Pu^{239} ratio was determined, these peaks were of negligible height. The source of those peaks has never been determined. Ions of masses 481 and 483 could, as doubly charged ions have produced the peaks at 240 1/2 and 241 1/2. A search of the region around mass number 480 showed no peaks.

Fifteen determinations of the abundance ratio Pu^{241}/Pu^{239}, made at three different temperatures of distillation of the sample, gave a value of $(4.12 \pm 0.08) \times 10^{-3}$. This value showed a slight tendency to increase with increasing temperature of the microfurnace. This effect is probably due to background from the extraneous 240 1/2 and 241 1/2 peaks and is almost within the limits of error of the measurement. Thus, the above value of the Pu^{241}/Pu^{239} ratio is probably high rather than low.

Curves of the oxide region run at the distillation temperatures at which the 240 1/2 and 241 1/2 peaks showed up, showed no evidence of peaks at the comparable positions, i.e., at mass positions 256 1/2 and 257 1/2.

All the above investigations were made from November 28 to December 6, 1945. About one-third of the sample had been distilled away to obtain these results.

The sample furnace was removed from the spectrometer tube and sealed off in an evacuated tube, preserving it for a later investigation. On February 28, 1946, this same sample (furnace and all) was sealed back into the spectrometer tube and a second series of determinations were made.

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Twelve determinations of the abundance ratio Pu^{240}/Pu^{239} gave a value of $(7.41 \pm 0.15) \times 10^{-2}$. Ten determinations of the ratio Pu^{241}/Pu^{239} yielded the value $(4.11 \pm 0.08) \times 10^{-3}$.

Immediately after the last set of ratios were determined, a curve was run without changing the furnace temperature. This curve did not show the 240 1/2 and 241 1/2 peaks in appreciable amounts. At higher temperatures, however, they were again seen.

In the last report of this series (LA-327), an investigationof a long series of extraneous peaks, which occurred in a sample of plutonium reduced to metal at Los Alamos, was reported. A series of investigations, reported in LA-327, showed that these peaks were caused by some material in the sample and not by some faulty mass spectrometric technique. Since this substance produced peaks at every mass position over the mass range 235 to 260, it was concluded that this substance must be hydrogenous.

Since that report was written, one further experiment was performed to attempt to shed further light on the character of this material. It seemed sensible to think that this material could be high-vacuum stopcock grease. The grease used by those people who had treated the sample after reduction was Celvasoene grease manufactured by the Distillation Products Co. in Rochester, New York.

Accordingly, the following experiment was performed. A clean micro-furnace was slightly contaminated with Celvasoene and sealed into the spectrometer tube. A spectrum of the 235 - 260 mass region was run with the temperature of the microfurnace very low (probably about 700 - 800°C). A spectrum was obtained showing peaks at every mass position over a wide range. However, the quantitative comparison with the spectrum obtained

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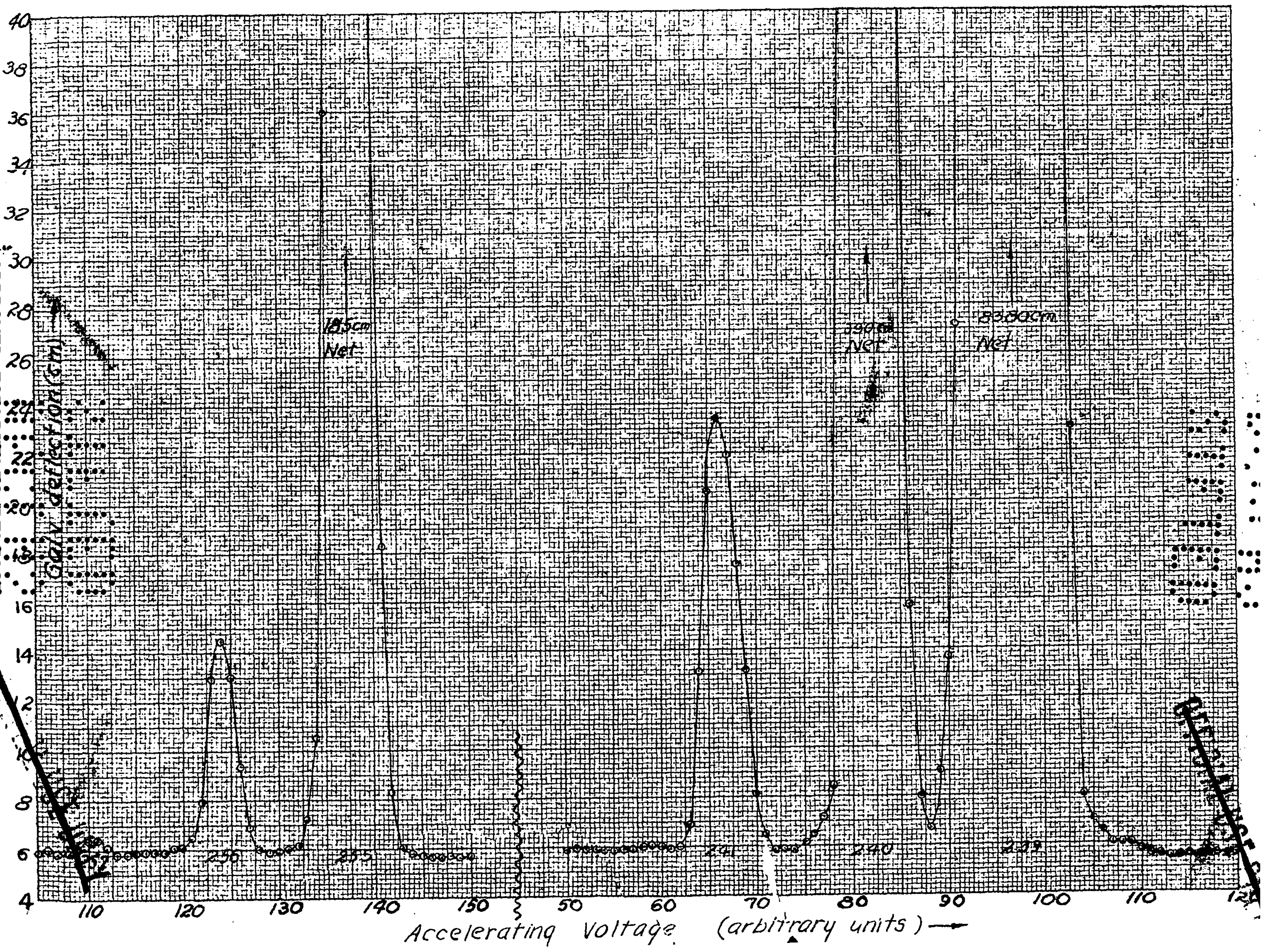
with the contaminated plutonium sample was not good, as was to be expected, because the conditions were far from identical in the two cases.

The conclusions to be drawn are (1) that, since a grease like Celvascene does produce peaks at every mass position in the range in which the unknown material falls, a grease of some kind could possibly be the unknown material, and (2) that the precise identity of this material is still not known.

SUMMARY OF DATA

Sample	Abundance Ratio $\frac{Pu^{240}}{Pu^{239}}$	Abundance Ratio $\frac{Pu^{241}}{Pu^{239}}$	Atomic % Pu^{239}	Atomic % Pu^{240}	Atomic % Pu^{241}	Spontaneous Fission Rate f/g hr of sample (source of data given in parenthesis)*	f/g hr. of Pu^{240}
Normal Clinton Plutonium	$(3.3 \pm 0.6) \times 10^{-4}$	---	99.97	0.033	---	---	
Clinton irradiated Plutonium (C-16 or CK-2A)	$(1.18 \pm 0.08) \times 10^{-3}$	---	99.88	0.118	---	1950 (±10%)(LA-490	$(1.65 \pm 0.25) \times 10^{6}$
Hanford shipment 41-H	$(1.11 \pm 0.02) \times 10^{-2}$	---	98.90	1.10	---	18,200 (±6%) (LA-490)	$(1.65 \pm 0.10) \times 10^{6}$
CW-1B	$(6.50 \pm 0.1) \times 10^{-3}$	$(8.1 \pm 0.6) \times 10^{-5}$	99.36	0.646	0.008	10,900 (±5%) (LA-490)	$(1.69 \pm 0.8) 10^{6}$
CW-2	$(4.65 \pm 0.05) \times 10^{-2}$	$(2.10 \pm 0.08) \times 10^{-3}$	95.37	4.43	0.200	70,000 (±3%) (LAMS-368)	$(1.59 \pm 0.06) \times 10^{6}$
CW-3	$(7.40 \pm 0.08) \times 10^{-2}$	$(4.12 \pm 0.08) \times 10^{-3}$	92.76	6.86	0.382	112,000 (±4%) (LAMS-383)	$(1.63 \pm 0.08) \times 10^{6}$
							ave f/g hr. $(1.64 \pm 0.06) \times 10^{6}$

*Note: Weighing of samples for spontaneous fission counting was done by two methods. (1) Alpha-counting and (2) fission counting in known neutron flux. The results of method (1) were corrected for alphas from Pu^{240} by use of the half-lives t 1/2 (Pu^{239}) = 24,400 yrs. and t 1/2 (Pu^{240}) = 6300 yrs. (Lams-293) The results of method (2) were corrected by using $\sigma_f (Pu^{240}) = \frac{\sigma_f (Pu^{239})}{10}$ (LA-444).



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