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Reaction Rate Calibration Techniques at ZPPR for ^{239}Pu Fission, ^{235}U Fission, ^{238}U Fission, and ^{238}U Capture

by

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Abstract

Reaction-rate calibration techniques used at ZPPR are described for ^{239}Pu fission, ^{235}U fission, ^{238}U fission and ^{238}U capture. In addition to these absolute reaction rates, calibration techniques are described for fission-rate ratios and the ratio of ^{238}U capture to ^{239}Pu fission. Uncertainty estimates are presented for all calibrations. Intercomparison measurements are reported which support the validity of the calibration techniques and their estimated uncertainties.

I. INTRODUCTION

In reaction rate measurements one is interested in knowing the number of atoms or fraction of atoms of a particular species which are formed or consumed in a given reaction. One common method for determining the number of atoms experiencing a reaction is to determine the number of reaction products by counting the number of gamma rays emitted by the reaction products. Because gamma-ray-counting systems have efficiencies less than unity, it is necessary to calibrate these systems by counting gamma rays from sources in which a known number of reactions have occurred. Thus, two counting measurements are required to make a reaction rate measurement; one with a sample in which a known number of reactions have occurred and a second sample in which the number of reactions which occurred is to be determined. In normal practice, a single calibration measurement will suffice to generate a calibration factor, ε, which can be used to determine the number of reactions which occurred in numerous, subsequent reaction rate measurements.

If C_γ is the number of counts recorded by the gamma-ray counting system from some reaction product, and if N is the number of reactions which occurred to produce these counts, then one can define the calibration factor ε_γ as:

$$\varepsilon_\gamma = \frac{(C_\gamma)_{calib}}{(N)_{calib}} \tag{1}$$

In a subsequent measurement one determines the number of reactions which occurred, $(N)_{meas}$ from the corresponding gamma-ray count and the calibration factor.

$$(N)_{meas} = \frac{(C_\gamma)_{meas}}{\varepsilon_\gamma} = \frac{(C_\gamma)_{meas}\ (N)_{calib}}{(C_\gamma)_{calib}} \tag{2}$$

The uncertainty in $(N)_{meas}$ will have contributions from both the calibration measurement and the subsequent sample counting. As shown in Eq. (2). $(N)_{meas}$ depends on the ratio of the two gamma-ray counts. Because these counts appear as a ratio, and because the two counts are obtained under nearly identical conditions, some uncertainties are correlated and therefore cancel. These uncertainty considerations are discussed in detail in later sections. All uncertainties quoted are at the 1σ level.

The following sections outline the reaction rate calibration procedures used at ZPPR for ^{239}Pu fission, ^{235}U fission, ^{238}U fission and ^{238}U capture reactions. The procedures used at the critical assemblies ZPR-6 and ZPR-9 at Argonne National Laboratory in Illinois are somewhat different than the procedures described here for the ZPPR assemblies in Idaho. The procedures for the reaction rate measurements and their calibrations were discussed previously in the ZPPR-4 breeding-ratio experiments.[1] Along with each calibration description an estimate is presented for the uncertainty associated with the calibration factor. These uncertainties in the calibration factor must be combined with uncertainties in the determination of $(C_\gamma)_{meas}$ in order to obtain the uncertainty in the reaction rate. A section is also included which presents the results of intercomparison measurements.

II. CALIBRATION METHODS AND STANDARDS

A. Fissionable and Fissile Foils and Deposits

The reaction rate calibration procedures used at ZPPR involve measurements with metal activation foils and with thin deposits on metal substrates. Foils of ^{239}Pu are 1.080 or 0.800 cm in diameter and nominally 0.013-cm-thick and are clad in 0.0025-cm-thick aluminum. These ^{239}Pu foils typically have masses of 150 or 90 mg. Both the ^{238}U and ^{235}U foils are 1.270 cm in diameter and nominally 0.013 cm thick, are coated in kel-F and are covered top and bottom with nominally 0.013-cm-thick aluminum discs. The masses of these metal foils are determined by weighing and the uncertainties in these masses is less than 0.1%. These uncertainties are not included in the overall calibration uncertainty because they are small compared to other sources of uncertainty in the calibration procedures. Uncertainties in the isotopic composition of the foils are also small.

An important component of the calibration measurements is the fission rate measured in back-to-back fission chambers. The fissionable and fissile materials in these chambers are thin deposits, 1.588 cm in diameter, 0.008-cm-thick stainless steel substrate discs. The fissionable and fissile materials are electroplated from solution and then fired in air to convert the metallic deposits to oxide.

The masses of these deposits are typically 100 μg for ^{235}U and ^{239}Pu and are about 250 μg for ^{238}U. The primary deposit mass standards are ^{235}U deposits. The masses of the ^{235}U standards are determined initially in a quantitative deposition. A weighed amount of ^{235}U is dissolved and a known fraction of this solution is removed by pipetting for use as the plating solution. The electroplating process removes approximately 99% of the uranium from the solution, and a fluorimetric technique is used to determine the ^{235}U mass remaining in solution. The mass of the deposit is then the difference between the intial and final ^{235}U masses in the plating solution. Pairs of deposits are then counted in a back-to-back fission chamber and the count rate ratios are compared to the mass ratios to verify the mass values. An additional mass verification can be made by dissolving one of the deposits and determining its mass by isotope dilution mass spectrometry. An alternative method for the initial mass determination is to measure the amount of fissile material in the plating solution before and after deposition using the isotope dilution method. The method of weighing, diluting, pipetting and fluorometric analysis results in an uncertainty of 1.2% in the mass of a ^{235}U deposit.

Once a mass standard is established for ^{235}U deposits, other fissile deposit masses are determined relative to ^{235}U in back-to-back fission chamber measurements in the thermal column of AFSR. (The AFSR thermal column has a cadmium ratio greater than 2000.) This method requires the knowledge of the thermal cross sections and g-factors for ^{235}U and for ^{239}Pu when ^{239}Pu deposit masses are determined.

In order to evaluate the uncertainty in deposit masses derived from the ^{235}U masses, it is necessary to estimate the uncertainty associated with the back-to-back fission chamber. A correction is applied to the observed countrates for fission fragment pulses lost below a predetermined bias level. The magnitude of the correction is 1% and the uncertainty assigned is 0.4%.

The thermal column back-to-back fission chamber method was used to determine the mass of a ^{239}Pu deposit used in reaction rate calibration measurements. The uncertainty in this ^{239}Pu deposit mass had the following components: (1) counting statistics in the back-to-back fission chamber 0.1%; (2) ^{239}Pu fission cross section 0.42%; (3) ^{239}Pu g-factor 0.28%; (4) ^{235}U fission cross section 0.31%; (5) ^{235}U g-factor 0.16%, (6) ^{235}U deposit mass 1.2%, and (7) back-to-back fission chamber correction 0.4%. These uncertainties combine to give an overall uncertainty in the ^{239}Pu deposit mass of 1.4%. The cross section and g-factor values are given in Table I.

TABLE I. Cross Sections and g-Factors used in Reaction Rate Calibrations*

Isotope	2200 m/s cross section, barns	g-factor
^{235}U (fission)	580.2 ± 0.31%	0.9766 ± 0.16%
^{238}U (capture)	2.720 ± 0.6%	1.0021 ± 0.2%
^{239}Pu (fission)	741.6 ± 0.42%	1.0548 ± 0.28%

*From Hanna G.C., Westcott C.H., Lemmel H.D., Leonard Jr. B.R., Story J.S., Attree P.M., Atomic Energy Review 7 (1969) No. 4 p. 3.

The thermal column method with a ^{235}U reference deposit was also used to determine the masses of other ^{235}U deposits. The uncertainty of such a ^{235}U deposit had the following components: (1) counting statistics in the back-to-back fission chamber 0.05%; (2) ^{235}U reference mass 1.2%; (3) back-to-back fission chamber correction 0.4%. These uncertainties combine to give an overall uncertainty in the ^{235}U deposit mass of 1.3%.

The deposits used for calibrating the ^{238}U fission rate are made of natural uranium. The natural-uranium deposit is simultaneously irradiated in the AFSR thermal column with a reference ^{235}U deposit. The relative count rates from the two deposits allow the ^{235}U mass in the natural-uranium deposit to be measured. This ^{235}U mass in the natural-uranium deposit and the ratio of the ^{238}U mass fraction to the ^{235}U mass fraction yield the ^{238}U mass in the natural-uranium deposit.

Counts in the back-to-back chambers can be obtained in a relatively short time by irradiating the chambers at low power for the ^{235}U deposit and at higher power for the natural-uranium deposit. Alternatively, any potential uncertainty in power ratio measurements can be eliminated by irradiating the chambers at low power and increasing the counting time for the natural-uranium deposit. Both approaches have been used successfully. The uncertainty in the mass of ^{238}U in the natural-uranium deposit has the following components: (1) counting statistics in the back-to-back chamber 0.3%; (2) isotopic composition of the deposit 0.8%; (3) reference ^{235}U deposit mass 1.2%; (4) back-to-back fission chamber correction 0.4%. These uncertainties combine to give an overall uncertainty in the ^{238}U deposit mass of 1.5%.

B. The Back-to-Back Fission Chamber

The back-to-back fission chambers used in reaction rate calibrations are thin-walled aluminum hemispheres approximately 3.56 cm in diameter. The detectors are of the flowing-gas type, and are shown in a side view and a top view in Fig. 1. The two fissile deposit substrate discs are mounted back-to-back on a lip at the chamber center. In some measurements one or two irradiation foils are placed between the backs of the deposits. These foils are centered under the fissile deposits.

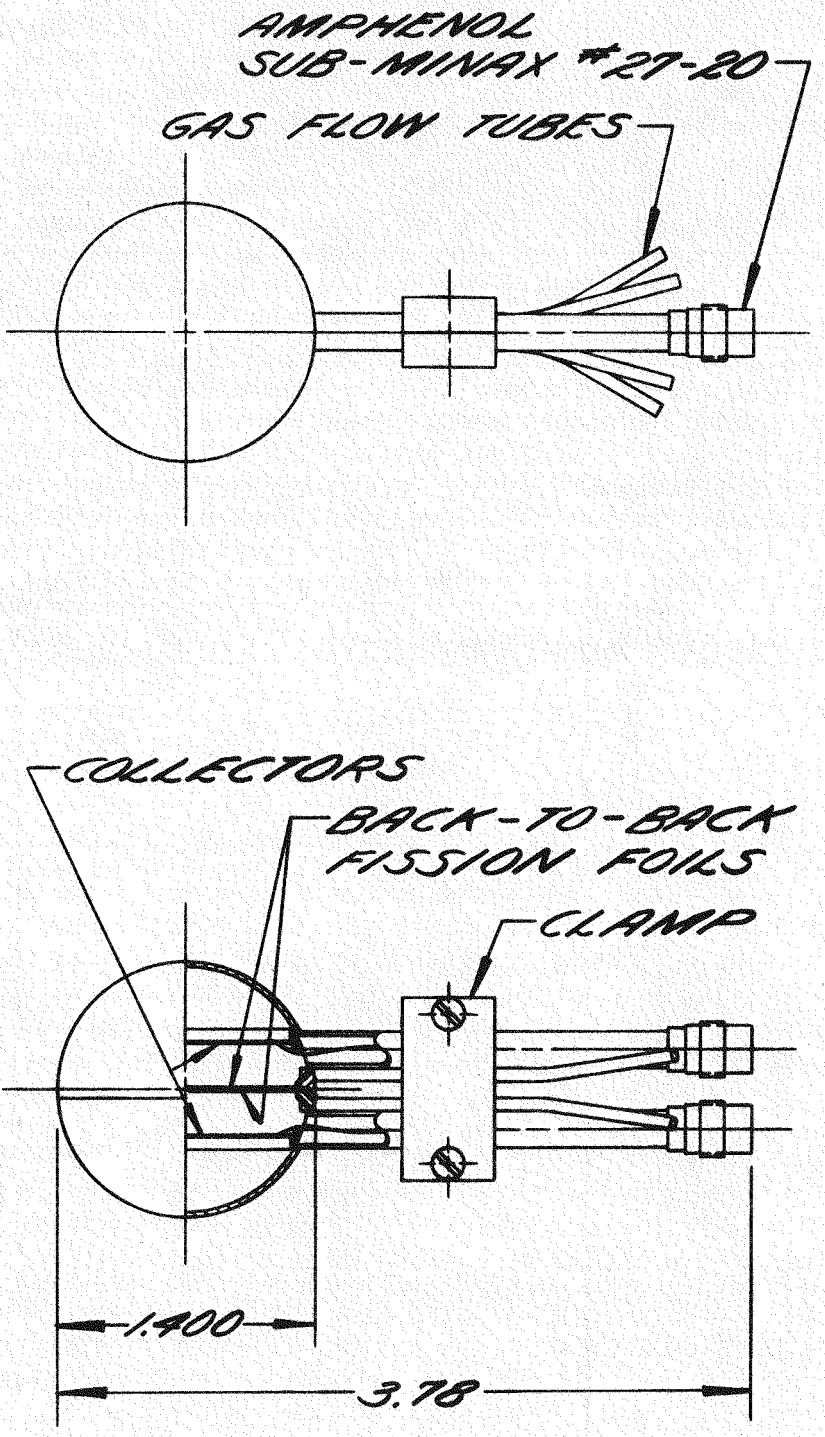

Fig. 1. Back-to-Back Fission Chamber.

An important assumption in using the back-to-back fission chambers is that both deposits see the same neutron flux and neutron spectrum. However, the chambers are irradiated in reactor environments in which flux gradients are known to exist. In order to minimize the effects of flux gradients it is best to keep the plane of the fissile deposit perpendicular to the direction of the flux gradient. This is not always possible because the counting-gas tubes and cables protruding from the chamber restrict the freedom of movement of the detector. Also of concern is the possibility that the neutron absorption and/or fission in activation foils between the deposits can itself cause a flux gradient. The effects of flux gradients can be detected by obtaining a countrate ratio, rotating the chamber 180° and then obtaining a second countrate ratio. If the two ratios are different, an average of the two values can be used. When no activation foils are present and the back-to-back chamber is in the AFSR thermal column, the difference between count rates for different chamber orientations is less than 0.3%. If a ^{238}U activation foil is inserted, this difference is about 0.5%.

The sensitivity to flux gradients and self-shielding effects of the back-to-back chamber with a ^{239}Pu activation foil between deposits was measured in a series of special experiments in ZPPR. Any shielding effects of the activation foil on the deposits and any neutron self-shielding effects within the activation foil were expected to be a function of the neutron spectrum. Thus if shielding or self-shielding effects are significant, the measured absolute ^{239}Pu fission rate calibration constant would be a function of reactor environment. In the series of special experiments, absolute ^{239}Pu and ^{235}U fission rate calibration constants were determined in different reactor environments characterized by different neutron spectra. These environments included inner core, outer core, blanket and control-rod-positions regions. No correlation was detected between calibration constant and reactor environment within the less than 2% uncertainties in individual constants. Because there was no statistically significant variation in the measured calibration constants with reactor environment, self-shielding corrections were not applied to back-to-back chamber data when ^{239}Pu or ^{235}U activation foils were present in the chambers during ZPPR irradiation. The case of ^{238}U deposits (in natural uranium) and uranium foils is less clear. For a ^{238}U deposit and a ^{238}U foil, no significant effect is expected. However, past practice was to simultaneously irradiate a ^{238}U foil and a ^{235}U foil in a back-to-back chamber containing a ^{235}U deposit and a natural-uranium deposit. There was some evidence that the fission neutrons from the ^{235}U foil may have induced a small (~ 1%) increase in the ^{238}U fission rate in the natural-uranium deposit. The present calibration procedure eliminates the ^{235}U foil, and experiments are planned to ascertain whether or not a ^{235}U foil has any effect on the observed ^{238}U fission rate. To date, no corrections have been applied to back-to-back chamber data for the effects of any of the foils on any of the deposits.

Because individual fission products are used to measure the number of fissions occurring in a foil, it is possible that variations in neutron spectrum could produce variations in the yields of these fission products for a constant number of fissions. Again, the lack of correlation between observed calibration constant and reactor location indicates that fission product yield does not vary significantly with reactor location although this effect may not be separable from other spectral effects. In addition, recent studies of the variation of fission product yields with neutron energy[2,3] indicate that differences in fission yields are less than 2% for the differences in neutron-energy spectrum observed for the various ZPPR regions.

C. Gamma-Ray Counting

This section describes the adjustments to the gamma-ray count data from irradiated foils which are used in the reaction rate calibrations. Also discussed are the uncertainties associated with the count data and the adjustments. All gamma spectra were collected with high-resolution Ge or Ge(Li) spectrometers. Details of the counting system are presented elsewhere[4,5].

For ^{235}U fission and ^{238}U fission, gamma ray peaks are analyzed for four fission products. For the case of ^{239}Pu fission, peaks are analyzed for three fission products. For the case of ^{238}U capture, a single peak is analyzed for the ^{239}Np gamma ray at 278 keV. Table II lists those gamma rays, their energies and their half lives. In addition, the 772 keV gamma ray from the ^{132}I fission product is sometimes also analyzed in the case of ^{239}Pu fission. These fission products are all in equilibrium with their precursors.

There are other fission-product gamma-ray peaks which overlap those peaks listed in Table II. These overlapping peaks are subtracted from the peaks of interest by the gamma-ray-data-processing computer program. The contribution from these overlapping peaks are normally less than 10% of the area of the peak of interest. The uncertainty associated with this overlap subtraction is less

TABLE II

Isotope	Half-Life, hr	Gamma-ray Energy, keV
^{239}Np	56.304 ± 0.17%	278
^{133}I	20.8 ± 0.48%	529
^{97}Nb	17.0 ± 1.18%	658
^{132}I	77.7 ± 1.03%	667
^{97m}Nb	17.0 ± 1.18%	743

than 10%. This uncertainty is not propagated when considering the overall uncertainty in the calibration factor because in the determination of experimental reaction rates, the number of gamma-ray counts in an experimental measurement is divided by the number of gamma-ray counts in the calibration. Because the gamma-ray counting procedure is the same for both the experimental measurement and the calibration, any systematic errors in the overlap subtraction process should be expected to cancel.

The number of gamma-ray counts in both experimental measurements and in calibrations are begun approximately 18 hours after the irradiations are completed. A correction must be applied to the number of fission product atoms to account for decay following irradiation. This decay correction factor is just $e^{-\lambda t_c}$ where t_c is the time between the end of the irradiation and the recording of the gamma-ray counts and λ is the decay constant for the species being counted. Gamma-ray counting generally continues until 70-84 hours after the end of the irradiation.

It is also necessary to correct the number of fission product atoms (or capture product atoms in the case of ^{238}U capture rate measurements) for decay during the irradiations. Irradiations typically last 0.5-12 hours. The

correction term for decay during irradiations is $\sum_{i}^{tr}(x_i\ \Delta t e^{-\lambda(tr-t_i)})$ where Δt is the data-taking time interval for reactor power, t_i is the time of some ith time interval, tr is the total irradiation time, and x_i is the reactor power during the ith time interval. During foil irradiations the gamma-ray data processing system monitors the reactor power. The output from a power monitor is integrated over a time interval Δt, which is typically 5 sec. At the end of a run this power history is punched on paper tape. In the case of ^{238}U capture it is also necessary to make a correction for the delay in the ^{239}Np decay caused by the 23.54-minute half-life of ^{239}U. A correction factor, $(1-\mu)$, is introduced where μ is the reduced half-life. That is,

$$\mu = \frac{(t_{1/2})_U}{(t_{1/2})_{Np} - (t_{1/2})_U} = 0.007$$

The reduced half-life correction factor is valid for cases in which there is at least a 5-hour delay between the end of the foil activation and the beginning of the gamma-ray count.

The decay constants for the various isotopes of interest have uncertainties of approximately 1% or less. Uncertainties are not included for the times tr, t_c or Δt or for the reactor power values x_i. Uncertainties in the decay correction terms are not propagated because, as in the case of the overlap subtraction, these uncertainties are expected to cancel because they appear in the reaction rate determinations as a ratio of counts for a rate measurement to counts for the calibration. An exception to the cancelling of decay-correction uncertainties occurs for the ^{243}Am calibration of the ^{238}U capture rate. This uncertainty is discussed in Section IV.B.

The largest single source of uncertainty in the number of gamma-ray counts is just that from counting statistics. In the calibration measurement this uncertainty is a approximately 0.5% for ^{239}Pu fission, 0.4% for ^{235}U fission, 1% for ^{238}U fission and 0.5% for ^{238}U capture. Experimental measurements normally have uncertainties due to counting statistics of 1% for ^{239}Pu fission, 0.7% for ^{235}U fission, 1.5% or greater (depending on location) for ^{238}U fission and 0.5% for ^{238}U capture. These counting statistic uncertainties include uncertainties associated with background subtraction. Background subtraction deals with the continuous portions of the gamma radiation from all fission products and is distinguished from peak overlap subtraction.

The second largest source of uncertainty in the number of gamma-ray counts is that associated with the positioning reproducibility of the irradiation foil with respect to the gamma-ray detector. This uncertainty has been determined by repetitive measurements with a single sample to be 0.3%.

Gamma-ray counts are corrected for gamma self-attenuation in the foils. For fission products, this correction is 1% or less and the correction is assigned a 10% uncertainty. The resulting uncertainty in the number of gamma-ray counts from fission products is therefore 0.1%. In the case of ^{238}U capture, the ^{239}Np gamma-ray self-attenuation correction is a approximately 7% and the resulting uncertainty in the number of gamma-ray counts from ^{238}U capture is 0.3%. Generally, these uncertainties in the gamma-ray self-attenuation-correction factors are not propagated because these factors appear as a ratio in the reaction rate expression. An exception is the case of ^{238}U capture

rates calibrated with the ^{243}Am source. In this case the uncertainty in the self-absorption correction factor is included in the reaction rate uncertainty.

It is not strictly necessary to include gamma-ray self attenuation factors or their uncertainties because the calibration factor determination does not distinguish between gamma-ray losses in the sample from gamma-ray losses due to the non-unity efficiency of the detector. However, because not all foils have exactly the same thickness, these self-absorption factors are generated for each foil. An additional correction may be needed if activation foils of non-standard dimensions are used. The correction in the number of counts for dimensional differences is less than 1% and the resulting uncertainty in the number of gamma-ray counts is less than 0.1%.

Calibration of the gamma-ray counting system by counting foils irradiated in back-to-back fission chambers is done only infrequently. Between calibrations involving back-to-back fission chambers, the efficiency of the system is monitored by counting gamma-ray sources of known strength. The sources primarily used are ^{137}Cs, ^{54}Mn, ^{60}Co and ^{243}Am. An additional set of sources which includes ^{125}Sb, ^{110}Ag, ^{139}Ce, ^{7}Be, ^{65}Zn, ^{113}Sn and ^{207}Bi is sometimes used. The purpose of this source counting is to ascertain that no significant change in efficiency has occurred. Occasionally small efficiency changes of up to 1.5% are noted, and the source counting measurement is used to generate a correction factor to the previously-measured efficiency. Such changes are attributed to changes in the Ge(Li) detectors themselves. The counting system gain (energy calibration) is verified using a ^{226}Ra source every time a new set of foils is counted.

D. Alpha Counting

Alpha counting is primarily used in the reaction rate calibration process to determine the activity of ^{243}Am deposits used in the ^{238}U capture and ^{238}U capture/^{239}Pu fission ratio calibrations. The submicrogram deposits of ^{243}Am are approximately 1.27 cm in diameter and are electroplated onto substrates. The ^{243}Am deposits normally contain some ^{241}Am impurity which requires that the alpha counting system have energy discrimination capabilities.

Alpha counting is normally performed in systems with well-defined source-detector geometry so that the detector is sensitive to a well-known fraction of the total disintegrations in the sample. Uncertainties due to geometry are typically 0.14%. Because deposits are very thin, self-absorption corrections are not required. Typical uncertainties due to counting statistics are 0.1% for ^{243}Am. The uncertainty in the ^{243}Am activity introduced by the correction for the presence of ^{241}Am depends on the fraction of ^{241}Am present, but was typically 0.2% for deposits used at ZPPR. Thus, the total uncertainty in the activity of the ^{243}Am deposits is estimated to be 0.3%. The ^{243}Am source is covered with 0.025 cm of mylar to prevent recoil losses of ^{239}Np when alpha counting is not in progress.

III. ABSOLUTE ^{239}Pu FISSION RATE CALIBRATION

A. Calibration Procedure

An absolute calibration for the ^{239}Pu fission rate can be made by counting on the gamma-ray counting system a reference ^{239}Pu foil activated in a back-to-back fission chamber in ZPPR. The back-to-back fission chamber also contains a ^{239}Pu deposit of known mass. Normally the back-to-back chamber also contains a ^{235}U deposit and a ^{235}U foil. During a portion of the reference foil irradiation, the number of fission events occurring in the deposit is monitored by the fission chamber. The number of fissions which occur in the reference activation foil is then determined from the relative masses of the activation foil and the deposit, from the number of fissions counted in the chamber, and from the ratio of the integrated power during the foil activation to the integrated power during the fission chamber count. Equation (3) describes this relationship between the number of fissions occurring in the activation foil and the number of fission events counted in the chamber.

$$(N_{af}^{f})_o = \frac{m_{af}^{49}}{m_{dep}^{49}} \frac{\sum_{i}^{tr} (x_i \Delta t)}{\sum_{i}^{t_{fc}} (x_i \Delta t)} C_{fc} = \frac{m_{af}^{49}}{m_{dep}^{49}} \tau C_{fc} \qquad (3)$$

where: $(N_{af}^{f})_o$ is the number of fissions which occur in the activation foil

m_{af}^{49} is the mass of the activation foil

m_{dep}^{49} is the mass of the ^{239}Pu deposit in the fission chamber

C_{fc} is the number of counts recorded by the fission chamber

$\sum_{i}^{tr} (x_i \Delta t)$ is the integrated power producing fission in the activation foil

$\sum_{i}^{t_{fc}} (x_i \Delta t)$ is the integrated power during the fission chamber counting measurement. The time of this measurement is t_{fc}.

τ is the integrated-power ratio.

After the reference ^{239}Pu foil irradiation is complete, the foil is removed from the fission chamber and gamma-ray counts are recorded for the fission products listed in Table I. The various correction factors are applied to the gamma-ray counts as described in Section II.C.

The calibration factor which relates the number of gamma-ray counts to the number of fissions occurring in a foil is then

$$\varepsilon_{\gamma}^{f} = \frac{C_{\gamma}^{f}}{(N_{af}^{f})_t} \qquad (4)$$

where ε_γ^f is the absolute fission calibration factor, C_γ^f is the number of gamma counts from the activation foil and $(N_{af}^f)_t$ is the number of fission product atoms remaining in the foil at the time at which the foil is counted. $(N_{af}^f)_t$ is related to $(N_{af}^f)_o$ by the expression

$$(N_{af}^f)_t = (N_{af}^f)_o \, e^{-\lambda^f t_c} \left(\sum_i^{tr} (x_i \Delta t) e^{-\lambda^f (tr - t_i)} \right)$$

substituting Eq. (3) for $(N_{af}^f)_o$, one obtains the expression for ε_γ^f.

$$\varepsilon_\gamma^f = \frac{C_\gamma^f \, (1+W_f) \, m_{dep}^{49}}{e^{-\lambda^f t_c} \sum_i^{tr} (x_i \Delta t e^{-\lambda^f (tr - t_i)}) \, m_{af}^{49}} \, \frac{1}{C_{fc}} \, \frac{1}{\tau} \tag{5}$$

where W_f is the self-absorption correction for gamma-rays from fission products and λ^f is the fission-product decay constant.

B. Uncertainties

There are two primary components to the overall uncertainty in ε_γ^f in Eq. (5). These two components are the mass of the ^{239}Pu deposit, m_{dep}^{49}, and the number of gamma-ray counts, C_γ^f. The uncertainty in m_{dep}^{49} was evaluated in Part II.A. and was assigned a value of ±1.4%. The uncertainty in C_γ^f has two components: (1) counting statistics, 0.5% and (2) repositioning uncertainty, 0.3%. These components combine to give a resulting uncertainty in C_γ^f of 0.6%.

The other terms in Eq. (5) are not assigned uncertainties. As explained in Section II.C., the decay correction factors are not included because they appear as ratios in the actual reaction rate determinations. The uncertainty in m_{af}^{49} is very small compared to other uncertainties and is not specifically included. The power ratio τ is not assigned any uncertainty because all the component uncertainties are believed to be small and all are believed to cancel when the power ratio is taken. Finally, no uncertainty is included for C_{fc} for ^{239}Pu and ^{235}U because all the uncertainties associated with the number of counts from the back-to-back fission chamber were included in the uncertainty for m_{dep}^{49}. Normally uncertainties due to counting statistics in the fission chamber are approximately 0.1%. However, in the measurement of ^{238}U fission rates, this counting statistic uncertainty becomes significantly larger than 0.1%, and it is necessary to include explicitly this counting uncertainty associated with C_{fc}.

For ^{239}Pu fission, the 0.6% uncertainty in C_γ^f and the 1.4% uncertainty in m_{dep}^{49} combine to give a total uncertainty in ε_γ^f of 1.5%.

IV. ABSOLUTE ^{235}U FISSION RATE CALIBRATION

The procedure for absolute ^{235}U fission rate calibration is identical to the procedure for the absolute ^{239}Pu fission rate. The back-to-back chamber irradiation of a ^{235}U foil and a ^{235}U deposit is done along with the ^{239}Pu foil and deposit. The expression for ε_γ^f for ^{235}U fission is identical to Eq. 5 except that m_{dep}^{49} and m_{af}^{49} become m_{dep}^{25} and m_{af}^{25}.

The uncertainties in the ^{235}U fission calibration are also analogous to those for ^{239}Pu. The uncertainty in m_{dep}^{25} is assigned a value of 1.3%. The uncertainty due to gamma counting statistics is 0.4% and the repositioning uncertainty is 0.3%. These uncertainties combine to give a total uncertainty in ε_γ^f for ^{235}U of 1.4%.

V. ABSOLUTE ^{238}U FISSION RATE CALIBRATION

The absolute ^{238}U fission rate calibration is performed by simultaneously irradiating a depleted-uranium foil, a natural uranium deposit, and a ^{235}U deposit in a back-to-back fission chamber in ZPPR. The number of fissions in the back-to-back chamber due to ^{238}U in the natural-uranium deposit is the difference between the total fission counts from the natural-uranium deposit and the fission counts due to ^{235}U in the natural-uranium deposit. The number of fission counts due to ^{235}U in the natural uranium is determined from the number of fission counts from the ^{235}U deposit and from the relative number of ^{235}U atoms in the ^{235}U and natural-uranium deposits. The ^{235}U contribution to fission counts from the natural-uranium deposit is typically 23% of the contribution from ^{238}U. In a typical measurement in which 50,000 total fission counts are obtained from the natural-uranium deposit, the uncertainty in fission-chamber counts due to ^{238}U fission is 0.7%. This uncertainty in C_{fc} for ^{238}U fission is larger than the uncertainty in C_{fc} for either ^{235}U or ^{239}Pu fission and is combined with other uncertainties.

The expression for ε_γ^f for ^{238}U is the same as Eq. 5 except that m_{dep}^{28} and m_{af}^{28} are substituted for m_{dep}^{49} and m_{af}^{49}. The components of the uncertainty in ε_γ^f for ^{238}U are: (1) 1.5% due to the uncertainty in m_{dep}^{28}, (2) 0.7% due to the uncertainty in C_{fc}, (3) 1% due to gamma-counting statistics and (4) 0.3% due to repositioning uncertainty. These components combine to give a resulting uncertainty in ε_γ^f of 2.0%.

VI. FISSION RATIOS

Because some uncertainty components are common to the various absolute fission rate calibrations, the overall uncertainty in calibration factor can be less for fission rate ratios than for individual absolute fission rates. The most important common uncertainty component is that for the mass of the ^{235}U reference deposit, and this uncertainty component does not contribute to the uncertainty in the calibration factors for fission rate ratios. The uncertainties associated with the calibrations for the fission ratios $^{235}U/^{239}Pu$, $^{238}U/^{239}Pu$ and $^{238}U/^{235}U$ are discussed below. For all fission rate ratios, the value of the calibration factor is just the ratio of the absolute calibration factors.

A. $^{235}U(n,f)/^{239}Pu(n,f)$

The calibration factor for both ^{235}U and ^{239}Pu absolute fission rates is given in Eq. 5. The ratio of Eq. 5 for ^{235}U to Eq. 5 for ^{239}Pu contains uncertainty components only from m^{49}_{dep}, m^{25}_{dep}, C^f_γ for ^{235}U and C^f_γ for ^{239}Pu. Uncertainties in C_{fc} for both ^{235}U and ^{239}Pu are small and are not included. The fissile deposit masses appear as the ratio $m^{25}_{dep}/m^{49}_{dep}$. But m^{49}_{dep} was determined in a thermal irradiation of the ^{235}U deposit and the ^{239}Pu deposit, so the only uncertainty components in the deposit mass ratio are the uncertainties associated with: (1) the ^{239}Pu fission cross section 0.42%, (2) the ^{239}Pu g-factor 0.28%, (3) the ^{235}U fission cross section 0.31% and (4) the ^{235}U g-factor 0.16%. In addition, it is necessary to include the 0.4% uncertainty in C^f_γ for ^{235}U and the 0.5% uncertainty in C^f_γ for ^{239}Pu. These six uncertainty components combine to give an overall uncertainty in the calibration factor for $^{235}U(nf,)/^{239}Pu(n,f)$ of 0.9%.

B. $^{238}U(n,f)/^{239}Pu(n,f)$

The ratio of Eq. 5 for $^{238}U(n,f)$ to Eq. 5 for $^{239}Pu(n,f)$ provides the calibration factor for the fission rate ratio. The uncertainty of the calibration factor is less than the uncertainty of the ratio of the two absolute calibration factors because several uncertainty components cancel in the ratio. The major term which cancels is the mass of the ^{235}U deposit. The uncertainty components which are included in the ratio are: (1) the uncertainty in C^f_γ for ^{238}U 1%, (2) the uncertainty in C^f_γ for ^{239}Pu 0.5%, (3) the uncertainty in the ^{238}U counts from the natural-uranium deposit irradiated in ZPPR 0.7%, (4) the uncertainty in fission chamber counts from the natural-uranium deposit irradiated in AFSR 0.3%, (5) the uncertainty in the mass fraction ratio for the natural-uranium deposit 0.8% and (6) the combined uncertainty from the ^{235}U and ^{239}Pu thermal cross sections and g-factors 0.6%. The resulting total uncertainty for the $^{238}U(n,f)/\ ^{239}Pu(n,f)$ calibration factor is 1.7%.

C. $^{238}U(n,f)/^{235}U(n,f)$

The uncertainty in the calibration factor for the $^{238}U(n,f)/^{235}U(n,f)$ ratio is 1.5%. The components to this uncertainty are: (1) the uncertainty in C^f_γ for ^{238}U 1%, (2) the uncertainty in C^f_γ for ^{235}U 0.4%, (3) the uncertainty in the ^{238}U counts from the natural-uranium deposit irradiated in ZPPR 0.7%,

(4) the uncertainty in fission chamber counts from the natural-uranium deposit irradiated in AFSR 0.3%, and (5) the uncertainty in the mass fraction ratio for the natural-uranium deposit 0.8%.

VII. ABSOLUTE ^{238}U CAPTURE RATE CALIBRATION

A. Thermal Column Measurements

1. Procedure

A calibration factor relating the number of gamma-ray counts from ^{239}Np to the number of ^{238}U captures occurring in an activation foil can be obtained in a simultaneous irradiation of a ^{239}Pu deposit and a ^{238}U foil in a back-to-back fission chamber in the AFSR thermal column. This procedure is similar to that described for the absolute ^{239}Pu fission rate calibration. Equation (6) gives the expression for $(N_{af}^{39})_o$, the number of ^{238}U captures occurring in the activation foil.

$$(N_{af}^{39})_o = \frac{m_{af}^{28}}{m_{dep}^{49}} \frac{239}{238} \frac{\sigma_{28}^c}{\sigma_{49}^f} \frac{g_{28}^c}{g_{49}^f} (C_{fc})_{th} (\tau)_{th} \qquad (6)$$

where the subscript th refers to a thermal-column irradiation and where σ_{28}^c and σ_{49}^f are the 2200m/sec capture and fission cross sections for ^{238}U and ^{239}Pu respectively. The corresponding g-factors are g_{28}^c and g_{49}^f. These cross sections, g-factors and their uncertainty were given in Table I. The mass of the ^{238}U foil and the ^{239}Pu deposit are, respectively m_{af}^{28} and m_{dep}^{49}. The other terms are as defined in Section III.A.

The capture rate calibration factor, ε_γ^c is defined in Eq. (7).

$$\varepsilon_\gamma^c = \frac{C_\gamma^c}{(N_{af}^{39})_t} \qquad (7)$$

where C_γ^c is the observed number of gamma-ray counts from ^{239}Np counted at some time t. The number of atoms of ^{239}Np produced, $(N_{af}^{39})_o$ must be corrected for the decay of ^{239}Np during and after irradiation in a manner analogous to that described in Section II.C. The decay correction factors are again $e^{-\lambda^{39} t_c}$ and

$$\sum_i^{tr} (x_i \, \Delta t e^{-\lambda^{39}(tr-t_i)})$$

where λ^{39} is now the decay constant for ^{239}Np.

A correction factor for gamma self-attenuation $(1+W_c)$ is also made to the number of counts as described in previous sections. The equation relating the number of ^{239}Np atoms at the time of counting, $(N_{af}^{39})_t$ to the number initially produced in the foil, $(N_{af}^{39})_o$ is:

$$(N_{af}^{39})_t = (1-\mu) (N_{af}^{39})_o e^{-\lambda^{39} t_c} \sum_i^{tr} (x_i \Delta t e^{-\lambda^{39} (tr-t_i)}) \qquad (8)$$

where μ was defined in Section II. Substituting Eq. (6) for $(N_{af}^{39})_o$ and correcting for self absorption one now can write

$$\varepsilon_\gamma^c = \frac{C_\gamma^c\,(1+W_c)\,m_{dep}^{49} \cdot 238 \cdot \sigma_{49}^f\, g_{49}^f}{(1-\mu)\,m_{af}^{28} \cdot 239 \cdot \sigma_{28}^c\; g_{28}^c\; (C_{fc})_{th}\,(\tau)_{th}\, e^{-\lambda^{39}t_c} \sum_i^{tr} (x_i \Delta t e^{-\lambda^{39}(tr-t_i)})} \quad (9)$$

2. Uncertainties

As in the case of the absolute ^{239}Pu fission rate, the largest sources of uncertainty in the absolute ^{238}U capture rate calibration factor are the mass of the ^{239}Pu deposit, m_{dep}^{49}, and the number of gamma-ray counts C_γ^c. The uncertainty in C_γ^c is approximately 0.5% due to counting statistics and 0.3% due to repositioning. These uncertainties combine to produce an overall uncertainty in C_γ^c of 0.6%. As discussed in Section III, no uncertainty is assigned to the decay correction factors, to the power factors, to C_{fc}, to m_{af}^{28}, to $(1+W_c)$ or to $(1-\mu)$. The terms σ_{49}^f and g_{49}^f also appear in the expression for m_{dep}^{49} and thus cancel from Eq. (9). Consequently no uncertainties are included for σ_{49}^f or g_{49}^f. The uncertainties in cross section and g-factor for ^{235}U are still included through the uncertainty in m_{dep}^{49}. The uncertainty in σ_{28}^c is 0.6% and the uncertainty in g_{28}^c is 0.2%. All these component uncertainties combine to give an overall uncertainty in ε_γ^c of 1.6%.

B. ^{243}Am Measurements

The ^{243}Am deposits described in Section II.D. can be used in the calibration for the absolute capture rate in ^{238}U. ^{243}Am decays by alpha emission to ^{239}Np with a half-life of approximately 7370 years. Because the ^{239}Np decays with a half-life of 2.35 days, the ^{239}Np quickly comes into equilibrium with the ^{243}Am. At equilibrium, the alpha activity of the ^{243}Am is equal to the beta-decay activity of the ^{239}Np that is,

$$\lambda^{53}\, N_{Am}^{53} = \lambda^{39}\, N_{Am}^{39} \quad (10)$$

where N_{Am}^{53} is the number of ^{243}Am atoms and N_{Am}^{39} is the number of ^{239}Np atoms in the ^{243}Am deposit. ^{243}Am alpha activity is determined as described in Section II.D.

The calibration factor for ^{238}U capture is just the ratio of the gamma-ray count rate (a measure of the ^{239}Np activity) to the ^{243}Am activity determined by alpha counting, i.e.,

$$\varepsilon_\gamma^c = (C_\gamma^{39})_{Am}/\lambda^{53}\, N_{Am}^{53} \quad (11)$$

The uncertainty of this calibration factor can be quite small because there are only three uncertainty components. The first component is the ^{239}Np counting statistics which can be made as small as 0.1% by long counting times. The 0.3% uncertainty due to sample positioning is also applied. The uncertainty in the ^{243}Am activity is 0.3% resulting in an overall uncertainty in the calibration factor of 0.4%. Minor corrections to the count rate are made to account for small differences in sample-detector geometry between the ^{243}Am deposit and activation foils. Uncertainties in these corrections are very small.

Because the ^{239}Np is in equilibrium with the ^{243}Am, the calibration factor ε_{γ}^{C} does not depend on λ^{39}. However, the experimental reaction rate data obtained from irradiated ^{238}U foils will depend on λ^{39}. The previous argument for ignoring uncertainties in decay corrections is not valid when the ^{243}Am calibration is used. However, the uncertainty appears in the number of gamma ray counts for the reaction rate measurement. The estimated uncertainty in λ^{39} is 0.17%. For a typical irradiation time of 4 hours and a decay time after irradiation of 84 hours, the uncertainty in C_{γ}^{C} is approximately 0.1%. The 84-hour decay time corresponds to the end of a normal gamma-ray-data-collection time.

In the past the ^{238}U capture rate calibration factor was also determined using the 279 keV gamma-ray from a known-activity source of ^{203}Hg. This procedure is no longer used because of relatively large uncertainties. However, calibration factors based on ^{203}Hg did agree within uncertainties with calibration factors determined by other methods.

VIII. THE ^{238}U CAPTURE/^{239}Pu FISSION RATE RATIO CALIBRATION

In addition to interest in the absolute reaction rates for ^{238}U capture and for ^{239}Pu fission, one is often interested in the ^{238}U capture to ^{239}Pu fission reaction rate ratio. A separate calibration for the ratio is useful because it is possible to eliminate the mass of the ^{239}Pu deposit from the expression for the calibration factor. Eliminating the ^{239}Pu deposit mass results in an uncertainty for the reaction rate ratio calibration factor which is less than the uncertainty obtained from the ratio of the two absolute reaction rate calibration factors.

A. ^{238}U Capture to ^{239}Pu Fission Ratio Calibration by Thermal Irradiation

1. Procedure

In the calibration of the capture-to-fission-rate ratio, the AFSR thermal column irradiation of a ^{239}Pu back-to-back fission chamber containing a ^{238}U foil is performed in the manner described in Section IV.A. The ^{238}U foil is removed and counted in the gamma-ray counting system in the usual way. In addition, the ^{239}Pu back-to-back fission chamber is used in the ZPPR reactor to irradiate a ^{239}Pu activation foil. This fission foil is then removed and counted in the gamma-ray counting system in the usual way. The capture-to-fission-rate ratio calibration constant $\varepsilon_\gamma^{c/f}$, is defined in Eq. 12.

$$\varepsilon_\gamma^{c/f} = \frac{C_\gamma^c / C_\gamma^f}{(N_{af}^{39})_t / (N_{af}^f)_t} = \frac{C_\gamma^c}{C_\gamma^f} \frac{(N_{af}^f)_t}{(N_{af}^{39})_t} \qquad (12)$$

where the symbols are as previously defined.

As before, $(N_{af}^{39})_t = (N_{af}^{39})_o \, e^{-\lambda^{39} t_c} \sum_i^{tr} (x_i \Delta t e^{-\lambda^{39}(tr-t_i)})$

and

$$(N_{af}^{39})_o = (1-\mu) \frac{m_{af}^{28}}{m_{dep}^{49}} \frac{239}{238} \frac{\sigma_{28}^c}{\sigma_{49}^f} \frac{g_{28}^c}{g_{49}^f} (C_{fc})_{th} (\tau)_{th}$$

also, $(N_{af}^f)_t = (N_{af}^f)_o \, e^{-\lambda^f t_c} \sum_i^{tr} (x_i \Delta t e^{-\lambda^f (tr-t_i)})$

and $(N_{af}^f)_o = \frac{m_{af}^{49}}{m_{dep}^{49}} C_{fc} \, \tau$

The gamma-ray count ratio is modified only by the self-absorption correction factors $(1+W_c)/(1+W_f)$. Thus, the expression for $\varepsilon_\gamma^{c/f}$ becomes

$$\varepsilon_\gamma^{c/f} = \frac{C_\gamma^c\,(1+W_c)}{C_\gamma^f\,(1+W_f)} \frac{\frac{m_{af}^{49}}{m_{dep}^{49}}}{\frac{m_{af}^{28}}{m_{dep}^{49}}} \frac{1}{(1-\mu)} \frac{238}{239} \frac{\sigma_{49}^f\, g_{49}^f}{\sigma_{28}^c\, g_{28}^c} \frac{\tau}{(\tau)_{th}} \frac{C_{fc}}{(C_{fc})_{th}}$$

$$\times \frac{e^{-\lambda^f t_c} \sum_i^{tr} (x_i \Delta t) e^{-\lambda^f (tr-t_i)})}{e^{-\lambda^{39} t_c} \sum_i^{tr} (x_i \Delta t) e^{-\lambda^{39} (tr-t_i)})} \quad (13)$$

obviously, the dependence on m_{dep}^{49} cancels.

2. Uncertainties

The components of the uncertainty in $\varepsilon_\gamma^{c/f}$ have all been discussed previously. The uncertainty in σ_{28}^c is 0.6% and in g_{28}^c is 0.2% while the uncertainty in σ_{49}^f is 0.42% and in g_{49}^f is 0.16%. The count rates C_γ^c and C_γ^f each have uncertainties of 0.5% due to counting statistics and 0.3% due to repositioning. Uncertainties are not assigned to the other terms as discussed previously. All the uncertainties included combine to give an overall calibration factor uncertainty of 1.1%.

B. ^{243}Am Measurements

An alternate and independent means of calibrating the capture-to-fission rate ratio may be found by reconsidering Eqs. (12), (7), and (11). The ^{243}Am deposit can provide the term $C_\gamma^c/(N_{af}^{39})_t$ in Eq. (12). By alpha counting of the ^{239}Pu back-to-back chamber deposit and the ^{243}Am deposit, the mass of the ^{239}Pu deposit can be expressed in terms of the ^{243}Am activity, the decay constant for ^{239}Pu, and the alpha-count-rate ratio. Finally, a simultaneous irradiation of the ^{239}Pu back-to-back chamber and a ^{239}Pu activation foil in ZPPR with subsequent gamma counting of the activation foil can give the relationship between the number of fissions occurring in the activation foil and the number of gamma counts recorded. This ^{243}Am-based capture-to-fission rate ratio calibration has the potential advantage of providing a calibration independent of thermal column measurements and consequently independent of thermal cross-sections and g-factors. This ^{243}Am calibration procedure has not yet been implemented, although independent determinations of ^{239}Pu chamber deposit masses by alpha counting have shown good agreement with masses determined by the methods described in Section II.

IX. INTERCOMPARISONS

The credibility of the calibration procedures described in the preceding sections and the credibility of the uncertainties assigned to these procedures has been strengthened by a series of intercomparison measurements. Most of these intercomparison measurements have been aimed at verification of the mass determinations of the fissile deposits used in the back-to-back fission chambers.

The first report of a large-scale intercomparison was that by Amundson in 1966[6] in which fission chamber deposits were compared among the ZPR-3 and AFSR reactors at ANL, the ZEBRA and VERA reactors in Great Britain, and the Flattop and Water-Boiler reactors at LASL. As one part of these measurements, a direct comparison was made of the masses assigned to fission chamber deposits by comparing fission rate ratios to assigned-mass ratios for ^{233}U, ^{235}U, and ^{239}Pu using the ZEBRA and AFSR reactors. The fission ratios agreed with the mass ratios from both reactors within the reported uncertainties. These comparison measurements are summarized in Table III.

TABLE III. Comparison of Chambers Containing the Same Fissile Isotope

Fissile Isotope	Reactor	Fission-Rate Ratio/ Assigned Mass Ratio
^{233}U	ZEBRA	1.000 ± 0.018
	AFSR	1.002 ± 0.018
^{235}U	ZEBRA	1.018 ± 0.017
	AFSR	1.005 ± 0.021
^{239}Pu	ZEBRA	1.010 ± 0.014
	AFSR	0.994 ± 0.018

The fission chambers used to obtain the data reported in Table III contained only a single deposit. One chamber from ZEBRA and one chamber from ANL, each containing a deposit of the same fissile isotope, were irradiated in each of the two reactors.

These same ZEBRA-AFSR comparison methods were used to generate fission ratios using fissile deposits from both facilities in both reactors. The fission rates for the isotopes ^{233}U and ^{239}Pu were compared to the fission rates for ^{235}U. The results of the comparisons are shown in Table IV. These fission rate ratios all agree within experimental uncertainty.

In a separate comparison, ANL fission chambers with ^{238}U, ^{235}U, ^{233}U, and ^{239}Pu deposits were irradiated in the LASL Flattop critical assembly and fission rates relative to ^{235}U were measured. These fission-rate ratios were then compared to ratios reported by LASL. These comparisons are summarized in Table V. Again the fission rate ratios agree within the quoted uncertainties.

TABLE IV. Fission-Rate Ratios from ZEBRA and ANL Chambers in ZEBRA and AFSR

Core	Isotopes	Chambers Used	Fission Ratios
ZEBRA 4A	$^{233}U/^{235}U$	ZEBRA 2/8	1.445 ± 0.021
		ANL 23/13B	1.455 ± 0.020
ZEBRA 4A	$^{239}Pu/^{235}U$	ZEBRA 14/8	0.974 ± 0.015
		ANL 33/13B	0.981 ± 0.014
ZEBRA 5A	$^{239}Pu/^{235}U$	ZEBRA 14/8	0.988 ± 0.015
		ANL 33/13B	1.006 ± 0.014
AFSR	$^{233}U/^{235}U$	ZEBRA 15/6	1.587 ± 0.017
		ANL k16/k5	1.583 ± 0.024
AFSR	$^{239}Pu/^{235}U$	ZEBRA 11/6	1.494 ± 0.016
		ANL k21/k5	1.501 ± 0.022

An additional comparison was made between two ANL ^{235}U deposits and a LASL ^{235}U deposit in the thermal column of the LASL Water Boiler reactor. The ratio of the observed countrate ratio to the reported mass ratio was 0.988 ± 0.012 for one ANL deposit and was 0.987 ± 0. 012 for the other deposit.

TABLE V. Fission-Rate Ratios Measured in Flattop Using ANL Fission Chambers and Deposits

Deposits	Isotopes	Measured Ratio	LASL Ratio Value
45/38	$^{238}U/^{235}U$	0.148 ± 0.002	0.146 ± 0.004
44/58	$^{233}U/^{235}U$	1.57 ± 0.02	1.63 ± 0.05
47/38	$^{239}Pu/^{235}U$	1.35 ± 0.02	1.40 ± 0.06

A subsequent set of extensive comparison measurements was made between ANL and the National Bureau of Standards (NBS)[7] and reported in 1978. In this comparison, several chamber deposits were prepared by standard means by ANL. These deposits were then counted in back-to-back fission chambers in the NBS reactor against standard NBS deposits and masses were assigned to the ANL-prepared deposits. Some of the ANL deposit masses were determined at NBS by relative alpha counting. These ANL-prepared deposits were next counted in back-to-back fission chambers in the thermal column of AFSR using ANL reference deposits to determine the masses of the new deposits. The results of the NBS and ANL back-to-back-chamber results are shown in the first three columns of Table VI. The next step in this comparison was alpha counting by ANL. The masses of these deposits as determined by alpha counting are shown in the fourth column of Table VI. The results in Table VI show agreement between the NBS and ANL mass values within the mass uncertainties quoted earlier. A bias of 0.7% was noted between the NBS and ANL results for ^{235}U masses.

Another intercomparison study was reported in 1979 by Poenitz, Meadows, and Armani.[8] This intercomparison included deposits form NBS, LASL, ANL, and the University of Michigan. Measurements included both alpha counting and fast-spectrum fission chamber measurements. The mass scales defined by the two LASL deposits, four ANL deposits, the NBS deposit and the University of Michigan

TABLE VI. Deposit Masses Determined by NBS and ANL

Deposit	Mass by Relative Fission Counting, μg		Mass by Relative Alpha Counting, μg
	NBS	ANL	ANL
N-U5-1	27.11 ± 0.38	27.05 ± 0.14	26.92 ± 0.13
N-U5-2	28.62 ± 0.39		28.51 ± 0.14
N-U5-3	53.1 ± 0.7	52.53 ± 0.28	52.67 ± 0.25
N-U5-4	50.8 ± 0.7		50.39 ± 0.24
N-U5-5	75.2 ± 1.1	74.99 ± 0.39	74.51 ± 0.36
N-U5-6	73.7 ± 1.0	73.08 ± 0.66	73.19 ± 0.36
N-P9-1	22.79 ± 0.30	23.24 ± 0.16	22.81 ± 0.13
N-P9-2	24.37 ± 0.32		24.52 ± 0.14
N-P9-3	43.0 ± 0.6	43.74 ± 0.31	43.38 ± 0.24
N-P9-4	45.2 ± 0.6		45.13 ± 0.26
N-P9-5	69.6 ± 0.9		70.17 ± 0.39
N-P9-6	74.5 ± 1.11	75.20 ± 0.75	75.00 ± 0.42
N-U8-5	80.4 ± 1.3		80.29
N-U8-6	84.0 ± 1.4		84.71

deposit all agreed within 1% while the mass scale corresponding to a fifth ANL deposit was 1.5% lower than the average of the five mass scales. This relatively low-mass scale deposit from ANL was not related to the deposits used in zero-power-reactor-reaction-rate measurements.

A comparison measurement between ANL-East and ANL-West was made in 1976. In this measurement, mass ratios were determined in the thermal column of AFSR for ANL-East and ANL-West ^{239}Pu deposits. The measured ratio was within 1% of the ratio of the assigned masses. The absolute mass of the ANL-West ^{239}Pu deposit was then determined by absolute alpha counting. Alpha spectroscopy was used to separate the contribution from ^{241}Am and the half-life for ^{239}Pu was 2.439×10^4 years. The absolute mass determined by alpha counting was within 0.4% of the assigned mass.

When considering the above corroborative measurements, it appears that the uncertainties used here for ^{235}U, ^{238}U and ^{239}Pu deposit masses, 1.2%, 1.5% and 1.4% respectively, are reasonable. In addition, these intercomparisons provide confidence in the measurement techniques themselves. The use of the back-to-back fission chambers, the corrections for the loss of fission fragments and for loss of signal pulses, and their uncertainties appear to be valid. The validity of the absolute alpha counting procedure also appears to be confirmed.

An important comparison of absolute fission rates was made between ANL-West and NBS and reported in 1980.[9] In this measurement a ^{235}U activation foil from ANL was irradiated in a NBS back-to-back fission chamber in the ISNF reactor at NBS. The fission rate determined by NBS from their deposit was 3415 ± 1.7% fission/ (sec. mg of ^{235}U). The activation foil was sent back to ANL-West for gamma-ray counting and the fission rate determined by ANL was 3440 ± 1.6% fission/ (sec. mg of ^{235}U). The difference was 0.73% which was well within the estimated uncertainties and which was consistent with the 0.7% bias between ANL and NBS mentioned earlier. Again this comparison measurement lends confidence in the measurement methods and their uncertainty estimates.

Recently an experiment was conducted to compare directly the ^{243}Am-calibrated Ge(Li) gamma-ray counting techniques for determining absolute ^{238}U capture rates with a mass spectrometric technique.[10] Identically-irradiated samples of ^{238}U were analyzed independently by the Ge(Li) technique and by a mass spectrometric technique at both ANL-East and ANL-West. For the samples analyzed at ANL-East, the mass spectrometric technique measured a ^{238}U capture rate 1.5% higher than the rate measured by the Ge(Li) technique. For the samples analyzed at ANL-West, the mass spectrometric technique measured a capture rate 2.4% higher than that measured by the Ge(Li) technique. The approximately 2% difference in absolute capture rates measured by these essentially independent techniques is larger than the approximately 0.5% uncertainty in the difference. The source of this difference has not yet been identified. However, the mass spectrometric comparison measurement does still lend credibility to the basic gamma-ray measurement and calibration technique used for ^{238}U capture.

Previously, there had been ANL-East and ANL-West comparisons of ^{243}Am activity measurements by alpha counting and by gamma-ray counting. Alpha-counting measurements agreed within 0.1% while gamma-ray measurements agreed within 0.3%. There had also been some previous ANL-East and ANL-West comparisons of absolute capture rates. One capture rate comparison conducted in 1979 involved ZPPR, the Applied Physics cross-section group, and the Illinois ZPR group. One set of irradiated ^{238}U foils was counted by both ZPPR and the Illinois ZPR group. A second set of irradiated ^{238}U foils was counted by both the Illinois ZPR group and the cross-section group. Both ZPPR and the cross-section group measured capture rates 3% higher than those determined by the Illinois ZPR group. This meant that ZPPR and the cross section group were in agreement with each other, but not with the Illinois ZPR group. Following the discovery of a discrepancy with two other ANL groups, the Illinois ZPR group increased the foil-detector distance in its gamma-ray counting system and a second intercomparison measurement was carred out in 1980. In this second intercomparison, ten standard ^{238}U foils were irradiated in ZPR-9 and subsequently counted by both the ZPPR and Illinois counting systems. The results from this comparison are shown in Table VII. The ^{243}Am calibration method was used. These values for the absolute capture rate are in good agreement within the predicted uncertainties.

TABLE VII. Comparison of Absolute ^{238}U Capture Rates

Foil Number	Absolute Capture Rate (Captures/gm (x10^9))		Difference, %
	ANL-East	ANL-West	
D-294	4.101 ± 0.57%	4.096 ± 0.53%	0.12
D-295	4.253 ± 0.58%	4.247 ± 0.53%	0.14
D-296	4.178 ± 0.61%	4.196 ± 0.54%	-0.43
D-285	4.370 ± 0.59%	4.384 ± 0.59%	-0.32
D-286	4.295 ± 0.55%	4.252 ± 0.59%	1.00
D-287	4.070 ± 0.56%	4.104 ± 0.53%	-0.84
D-289	3.769 ± 0.61%	3.782 ± 0.55%	-0.34
D-290	3.380 ± 0.61%	3.352 ± 0.55%	0.83
D-291	2.816 ± 0.59%	2.852 ± 0.56%	-1.28
D-293	2.372 ± 0.68%	2.346 ± 0.58%	1.10

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