Reducing Uncertainty in Nuclear Data

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Good detective work, combined with theory, experiments, and Bayesian analysis, has reduced by an order of magnitude the uncertainties in the evaluated rate of neutron-induced fission. That reduction allows more accurate simulation of weapon performance. Similarly, more accurate determination of neutron reactions on radiochemical neutron detectors has increased the capability to evaluate the results of past nuclear tests. In both instances, integral experiments with the critical assembly Jezebel are playing an invaluable role. Jezebel and Godiva are the infamous "unclad ladies" from the 1950s. Pictured at left, Jezebel consists of three components of a plutonium sphere that, when brought together, form a critical mass. Unclad, or not encased in neutron reflectors, Jezebel still can support a fast chain reaction with a hard neutron spectrum, characteristic of various nuclear devices.

Weapons performance depends directly on the rates of nuclear reactions, among which the neutron-induced fission chain reaction, shown schematically in the background on the opposite page, is one of the most important. The rates of neutron-induced fission and other neutron-induced nuclear reactions have been measured in numerous experiments. In this article, we describe a project to assess and reduce uncertainties in those basic reaction rates and thereby increase confidence in the predictions of Los Alamos weapons simulation codes (see the box "Uncertainty Quantification for Weapons Certification"). The rate of a nuclear reaction, or more precisely, the cross section for an incident particle to collide and interact with a nucleus[1], varies with the energy of the incident particle. For that reason, cross sections are typically measured at specific incident energies, and the measured values serve as input to the simulation codes. Any uncertainties in those energy-specific, or differential, cross sections translate into uncertainties in the prediction of the overall yield (total energy released) of a nuclear device and other "integral" quantities, so called because they result from the sum of repeated occurrences of the nuclear reaction over a range of incident energies and, in some cases, over the volume of the nuclear material. We present work on reducing uncertainties in two cross sections, both describing neutron-induced processes that are significant for weapon certification: the plutonium fission cross section (see Figure 1), which determines neutron multiplication in a plutonium fission chain reaction, and the cross section for iridium-193 to become the isomer iridium-193m (a long-lived excited state) through neutron inelastic scattering.[2] That process ($^{193}\text{Ir} + n \rightarrow {}^{193m}\text{Ir} + n'$) has played an important role in diagnosing weapons performance in past underground nuclear tests.

Our work on reducing fission data uncertainties for weapon certification is having an impact on other nuclear technologies. The GEN-IV nuclear reactor program is one such example. This program is exploring several future reactor concepts: more complete burnup of nuclear fuel, proliferation-resistant fuel cycles, and using the reactor as a "waste burner" to transmute long-lived radioactive nuclei into short-lived ones. When the long-time behavior of a GEN-IV reactor was simulated taking into account the best nuclear data available, the known uncertainties in the fission rates led to significant uncertainties in some of the key performance quantities such as nuclear criticality and transmutation rates. Both depend heavily on the fission rates for uranium, plutonium, and several minor actinides (neptunium, americium, and curium). On the basis of this finding, the Advanced Fuel Cycle Initiative Program at Los Alamos is supporting experimental and theoretical research to improve the highest-priority nuclear cross sections—particularly those of the minor actinides that are not currently understood.

Fission cross sections also matter to the nuclear-powered space mission to study Jupiter's moons. With the Laboratory's help, NASA is designing a compact nuclear reactor that will use highly enriched uranium (HEU) to power the plasma thrust engine. The energy output, criticality, radiation environment, and other important features of this reactor are predicted with radiation transport codes that simulate the production of neutrons by the fission process and their subsequent movement and participation in fission and other nuclear interactions. Even though the mission to Jupiter would be unmanned, a safe launch is most important; at the same time, we must also be able to guarantee that, if a crash were to occur, the probability of a criticality accident would be negligible. This project, therefore, also needs estimates of fission cross-section uncertainties—in this case, uranium-235 fission—to guide design of the space reactor.

Both statistical analyses of data

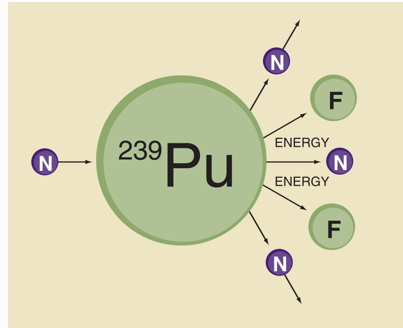


Figure 1. Schematic of Neutron-Induced Fission of Plutonium-239
This artist's conception of the fission process (as well as the simplified fission chain reaction in the background on the opposite page) shows an incoming neutron (purple) being absorbed by a plutonium-239 nucleus, which causes the nucleus to split into two 'fission' fragments (small green circles) and release several neutrons (purple). In reality, the nucleus first splits into two highly excited fragments and then each fragment releases one or more neutrons. The resulting fission fragments are typically radioactive nuclei and sometimes release additional ('delayed') neutrons. Both the 'prompt' and delayed neutrons can induce fission in nearby plutonium-239 nuclei, causing a fission chain reaction.

[1] A nuclear collision cross section $\sigma(E)$ measures the probability for an incident particle of energy E, say a neutron n, to collide and interact with or scatter from a nucleus N and produce some final state.

[2] In inelastic neutron scattering, the incident neutron n transfers energy to the nucleus N and leaves with less energy. That process ($N + n \rightarrow N + n'$) is denoted (n,n'), where the left neutron is incoming, the right neutron is outgoing, and the prime indicates that the outgoing neutron has a different energy than the incoming one.

Uncertainty Quantification for Weapons Certification

Since the end of nuclear testing, the Department of Energy has focused on developing a set of weapons simulation codes that more accurately model weapon explosions. This Advanced Simulation and Computing (ASC) Program has several objectives: creating simulation codes that implement more-accurate algorithms for solving the relevant hydrodynamics and radiation transport equations, building some of the fastest computers in the world on which to run these codes, and developing improved materials and physics models and data for "high-fidelity" weapons simulations. Such new simulation codes are needed to certify the safety and reliability of the U.S. stockpile and to answer questions about aging components in stockpiled weapons.

Quantification of the margins-and-uncertainties (QMU) concept has been adopted as the framework within which certification is performed. At each critical stage in the sequence of a weapon explosion, researchers in the Applied Physics Division at the Laboratory assess margins for certain physical quantities that enable the weapon to perform reliably. The QMU process formalizes the considerations and assumptions that go into modeling a weapon's performance and assessing whether it will perform correctly. A component of QMU is uncertainty quantification, whereby we determine how uncertainties in the underlying physics models and data impact the accuracy of full simulation results for weapons. It is in this context that we are assessing the accuracy of the plutonium fission cross-section data.

from past differential measurements and new state-of-the-art differential measurements at the Los Alamos Neutron Science Center (LANSCE) play a crucial role in allowing us to reduce cross-section uncertainties. More surprising, perhaps, is that small-scale integral experiments performed at the Los Alamos Critical Experiment Facility (LACEF) are having a huge impact in the validation of nuclear data used in weapons codes, as well as in reducing data uncertainties (see Figure 2). In the case of plutonium fission, for example, these criticality experiments have led to a factor of 10 reduction in the predicted fission process uncertainties,[3] as is discussed in more detail below.

As the name implies, a criticality experiment entails very careful assembling of a radioactive target made from special nuclear materials (plutonium, uranium-235 and -238, and other fissile materials) into a critical mass, that is, one that creates a self-sustaining fission chain reaction and a flux of neutrons with energies typical of fission. In fact, the energy spectra of the neutrons within the various assemblies at LACEF have been precisely determined through a combination of theory, simulation with radiation transport codes, and experiment. Thus, despite being integral experiments involving a wide spectrum of neutron energies and very large numbers of fission reactions occurring over a short period, critical-assembly experiments are well-characterized static nuclear physics experiments from which basic cross-section data can be inferred. In contrast, archival data from past Nevada underground nuclear tests were obtained from much more complicated integrated experiments involving hydrodynamics and other phenomena, in addition to nuclear physics.

Over the last few decades, nuclear criticality experiments have been used not only to reduce uncertainties in evaluated nuclear data libraries but also to validate the radiation (neutron and gamma-ray) transport methods used in our particle transport codes for static nuclear devices. One such code is the widely used Monte Carlo *N*-Particle Transport Code (MCNP). Developed by the Diagnostic Methods Group at Los Alamos, MCNP has become the international standard Monte Carlo code for simulating neutron transport and criticality in reactor applications and nuclear criticality safety studies. Nuclear criticality benchmark experiments developed at LACEF produce neutrons with a wide range of energy spectra: Some experiments mimic the highly thermalized systems of standard reactors, producing slow neutrons with an average energy of 0.025 electron volt (or soft neutron spectra); other experiments at the opposite extreme produce fast neutrons with an average energy of 1 to 2 million electron volts (MeV), or hard neutron spectra. The fast critical assemblies at LACEF are particularly relevant for validating our cross-section databases for weapons research because they produce a fast chain reaction (involving energetic neutrons). The Jezebel fast assembly is a critical mass of plutonium with no neutron reflectors, or cladding, the Godiva assembly is another 'unclad' assembly containing a critical mass of HEU, and the Flattop assemblies include cores of plutonium or HEU made critical with reflector materials.

[3] So-called "evaluated" nuclear data result from analyzing all available experiments, resolving discrepancies, and determining both the values and the uncertainties. They are kept in libraries known as ENDF for evaluated nuclear data files.

The two examples discussed below use fast critical-assembly measurements in different ways. In the case of plutonium, it is a precise measurement of the plutonium critical mass that allows us to accurately validate (and reduce the uncertainties on) the plutonium neutron-induced fission cross section, in part because our radiation transport methods in the MCNP code are so accurate. In the case of iridium, samples of iridium are placed at different locations within the critical assembly, and each is irradiated by a different spectrum of neutrons characteristic of its location within the assembly. The neutrons at different locations have not only different distributions of energies but also different mean energies. Thus, measuring iridium reaction rates within different parts of the assembly provides an important validation of the iridium cross sections at different average neutron energies.

Neutron-Induced Fission Cross Section of Plutonium

The neutron-induced fission cross section of plutonium-239 represents the probability that, when a single neutron hits a target nucleus of plutonium-239, the composite system of target plus neutron breaks apart, usually into two smaller nuclei fragments, $n + {}^{239}\mathrm{Pu} \rightarrow$ fission fragments. This probability naturally depends on the kinetic energy of the incident neutron and is therefore represented as a two-dimensional curve of cross section vs neutron energy (see Figure 3). To convert this probability into a rough estimate of the number of plutonium-239 fissions occurring in a real application—for example, in the core of a nuclear reactor—over a given period or in a critical assembly experiment, this cross section averaged over the neutron energies is multiplied by the neutron fluence (the neutron flux integrated over the relevant time).

Figure 2. The Los Alamos Critical Assembly Facility as Seen through an Anasazi Cave

Statistical Analysis of Experimental Data. The theory of nuclear fission has advanced considerably over the last fifty years, and especially within the last decade as high-performance computers made complex calculations feasible (see references by Peter Möller at the end of this article). Nevertheless, theoretical predictions of the neutron-induced fission cross section remain too imprecise for practical calculations of real systems. Experimental measurements of the cross section must therefore be relied on, and the cross section at most incident neutron energies is typically known to about 2 percent accuracy. Until now, however, the fission cross section for incoming neutrons of energies just below 14 MeV was known to only 4 percent accuracy. That deficiency motivated a significant effort to reanalyze the cross-section data from numerous (sometimes discrepant) experiments. We applied statistical methods to evaluate the cross-section data, assessed the resulting uncertainties, and were able to reduce uncertainties considerably.

An experiment typically yields a numerical value of a physical observable, which in turn is related either directly or indirectly to the physical quantity we are interested in. Of course, no experiment is perfect, and information on the uncertainties associated with the measured value is essential for judging the validity of the result. Uncertainties come from multiple sources but are commonly classified into two categories: statistical and systematic. Statistical uncertainties follow the simple $1/\sqrt{N}$ rule; that is, if the same experiment is repeated N times, the statistical uncertainty of the measured value will be proportional to $1/\sqrt{N}$. In the limit of an infinite number of identical experiments, this uncertainty would be null. Such uncertainties reflect inherent fluctuations in the measurement itself, and for a large number of repeated experiments, the measurement fluctuations average to zero.

Systematic uncertainties include all uncertainties other than statistical ones and, unlike the latter type, cannot be indefinitely reduced by repetition of the same experiment. Examples of systematic uncertainties will be given later for the plutonium-239 fission cross section. From the point of view of data analysis, systematic uncertainties define a lower limit for the accuracy of a given experimental setup. This fact alone justifies using different experimental setups to measure the same quantity. Because the sources of systematic errors differ from one experimental setup to another, differences in the results from different setups provide a clue on ways to go beyond the lower limits imposed by each individual experiment. By performing a statistical analysis on data from not only one but several experiments aimed at measuring or inferring the same physical quantity, it is possible to quote a value with an uncertainty smaller than the one of each individual experimental result.

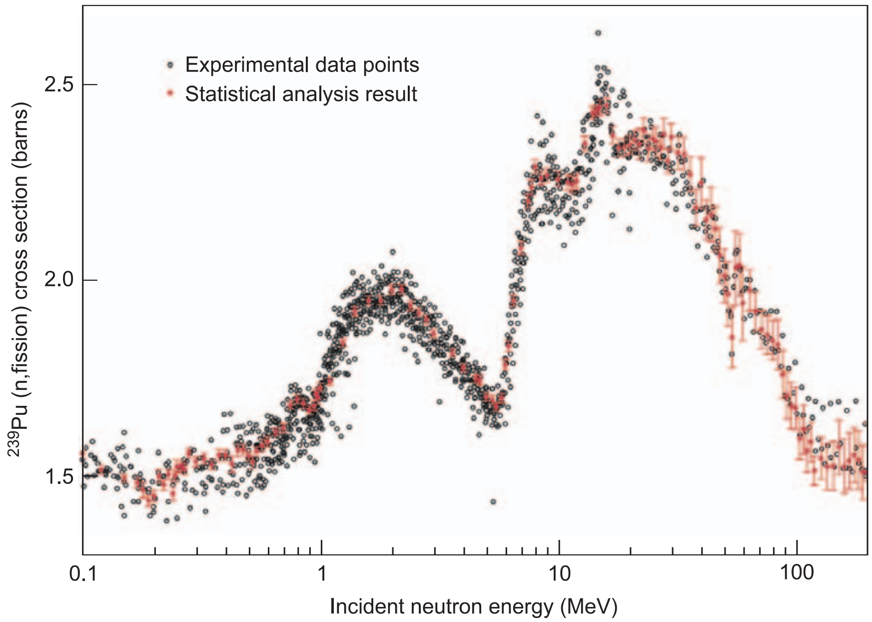


Figure 3. Reducing Uncertainties with Bayesian Statistical Analysis
The neutron-induced fission cross section of plutonium-239 has been measured numerous times over the incident-neutron energy range plotted here. The black dots represent the experimental data from many laboratories (including Los Alamos), which originate either from a direct measurement of the cross section or from a ratio measurement to the well-known neutron-induced fission cross section of uranium-235. (For figure clarity, we did not display the experimental error bars.) The spread of experimental data is a simple indicator of how well the cross section is known. The result of our Bayesian analysis study is shown in red dots, along with the resulting standard deviations. This figure explicitly demonstrates how a Bayesian statistical analysis can help reduce the uncertainties on our knowledge of this important cross section. At higher energies, the error bars tend to increase because two discrepant data sets are present.

In 1763, the work of Reverend Thomas Bayes on inference logic was published posthumously. Based on the theory of conditional probabilities, Bayes' theorem provides a logical and mathematically sound framework to update knowledge in view of new evidence. This concept is paramount in many areas of science and even more generally in any field of study that involves learning algorithms. Simply stated, Bayes' theorem reads

$$\mathcal{P}(\mathcal{H}|\mathcal{D},\mathcal{I}) \propto \mathcal{P}(\mathcal{D}|\mathcal{H}) \times \mathcal{P}(\mathcal{H}|\mathcal{I})$$
$$\text{Posterior} \propto \text{Likelihood} \times \text{Prior} \quad .$$

The term $\mathcal{P}(\mathcal{H}|\mathcal{I})$, or prior, answers the question, "how probable is the hypothesis $\mathcal{H}$, given the information known prior to the experiment?" In other words, the prior represents the state of our knowledge of (or belief in) the hypothesis $\mathcal{H}$ before the new information, in the form of the data $\mathcal{D}$, is included. The prior is multiplied by $\mathcal{P}(\mathcal{D}|\mathcal{H})$, the likelihood function, which quantifies how important the new data $\mathcal{D}$ are to our overall knowledge of the hypothesis $\mathcal{H}$. The likelihood function answers the question, "how probable is the observation of data $\mathcal{D}$ if the hypothesis $\mathcal{H}$ were actually true?" It provides the central and fundamental link between our prior knowledge and the posterior function $\mathcal{P}(\mathcal{H}|\mathcal{D},\mathcal{I})$, which answers the question, how probable is $\mathcal{H}$, now that we know both $\mathcal{D}$ and $\mathcal{I}$? In other words, the posterior measures the degree of confidence in $\mathcal{H}$ after the new data are taken into account.

Because a Bayesian analysis explicitly contains the concept of a prior knowledge, concerns have been raised about the subjectivity of such an approach, as opposed to more traditional statistical-analysis techniques. A Bayesian analysis is inherently a recursive process, in which information is integrated step by step. This means that the first step relies on a prior that is not based on any real information. When data are scarce, the result of the analysis can be distorted according to the specific choice made for the prior. This type of analysis appears to be in stark contrast with more traditional statistical analyses that are based on only real data. However, the contrast is only apparent, and the supposed flaw in the Bayesian approach seems to be only semantic. In any case, this issue is not relevant to our study: The number of data sets on the neutron-induced fission cross section of plutonium-239 is sufficiently large that the result of our analysis is insensitive to the choice of a particular prior.

The presence of this large data set could also lead us to think that much is known on this particular cross section and that there is no need to investigate further. The truth is not quite that simple. First, it is not uncommon to find discrepant experimental results, that is, results with error bars that do not overlap. Experimental data points for the plutonium-239 fission cross section are shown in Figure 3, illustrating how large the scattering in experimental results can be. Second, information on the uncertainties (and their sources) associated with a given data set is often only partially given, and for some (mostly older) experiments no information is available. As a result, our evaluation is all the more difficult. Finally, whereas most experimental results will be accurate at a

3 to 10 percent level, some important applications that need the plutonium-239 fission cross section require an accuracy closer to 1 to 2 percent. As mentioned earlier, a statistical analysis, Bayesian or otherwise, can help to more precisely determine the fission cross section.

We used a standard Bayesian approach to evaluate the plutonium-239 fission cross section from incident-neutron energies between 0.1 and 150 MeV. This energy range corresponds to a region where the cross section is a fairly smooth function of the incident energy (no resonances) and where the fission channel is dominant compared with other competing neutron-induced processes such as neutron capture, inelastic neutron scattering, and (*n*,2*n*) reactions, in which a nucleus absorbs the incoming neutron and promptly emits two.

Although the mathematical toolbox to evaluate the fission cross-section data was in place, inherent in this task was the need to reconstruct the uncertainties and correlations of important unpublished fission measurements that were performed (often many years ago) at numerous facilities around the world. This need required detective work.

In many cases, we were almost completely dependent upon the expertise of senior nuclear-data experimentalists and theorists, many of whom have retired or are close to retirement. These experts have in-depth knowledge of measurements made decades ago and a good (sometimes intuitive!) understanding of which experimentalists and facilities are most reliable.

Sources of experimental uncertainties are numerous and varied, depending on the particular experimental facility, detectors, and measurement and analysis techniques employed. In addition, the measured observable is often some function of the physical quantity of interest rather than the quantity itself. To determine the fission cross-section, for example, one measures the number of fissions produced during neutron irradiation of the target, which is proportional to fission cross section times the neutron fluence (defined as the neutron flux integrated over time). The neutron fluence is quite difficult to measure precisely and therefore introduces a large uncertainty into the results. Consequently, many experiments do not measure the plutonium fission cross section directly. Instead, they measure the ratio of the plutonium-239 to the uranium-235 fission cross section. By measuring that ratio, they eliminate the dependence on the neutron fluence and thus a large source of uncertainty. But a ratio is not a cross section. To come back to the quantity of interest, the experimental result needs to be multiplied by an 'evaluation' of the uranium-235 fission cross section, that is, a carefully determined result along with the uncertainties. Uncertainties on this cross section will then act upon the uncertainties on the plutonium-239 fission cross section in a highly correlated manner.

The neutron-induced fission cross section of uranium-235 is denoted as a standard cross section, one that experimentalists can use with confidence to renormalize their results (that is, convert measured ratios into cross sections) because it is a smooth function over a certain energy range and known very accurately. However, our knowledge of this cross section has changed over the years, by up to 2 percent in some energy regions. These differences are large enough that we have had to renormalize, according to current standard values, all the older experimental results obtained with standards known at the time in order to make a direct comparison of experimental data.

There are numerous other sources of uncertainties that must be analyzed. Over the years, different types of detectors have been used to measure or infer neutron-induced fission cross sections: A fission chamber that detects one (or sometimes the two) fission fragment(s), a proton telescope that uses the (*n*,*p*) reaction to estimate the neutron fluence, a time-of-flight (TOF) measure of the neutron incident energy, and others. Depending on the particular experimental setup, we have attempted to estimate the uncertainties associated with a given measured result after the fact, even though the experimentalist has recorded only partial or no information regarding error sources. In many cases, experimentalists have reported only statistical errors although a correct estimation of the systematic uncertainties is necessary for obtaining a quality result. Sometimes, we can fill in some of that information. For example, if two experiments have been performed in the same institute, they often use the same neutron source and target samples, in which case we include correlations between the two results in our analysis. In addition, documentation of the experimental details in one case can help us infer the values of uncertainties in the other.

Uncertainties may also exhibit an energy dependence. For example, if a detector efficiency is known to a certain accuracy within a given energy range, that accuracy defines some correlation among the results obtained with that detector within the specific energy range.

All these nonstatistical uncertainties cannot be described by simple standard deviations, but by correlations between fission cross sections at different energies. Correlations in the fission cross section are best represented by the so-called covariance matrix, whose diagonal and off-diagonal elements represent the standard deviations and the correlations, respectively. The off-diagonal elements play a key role in our statistical

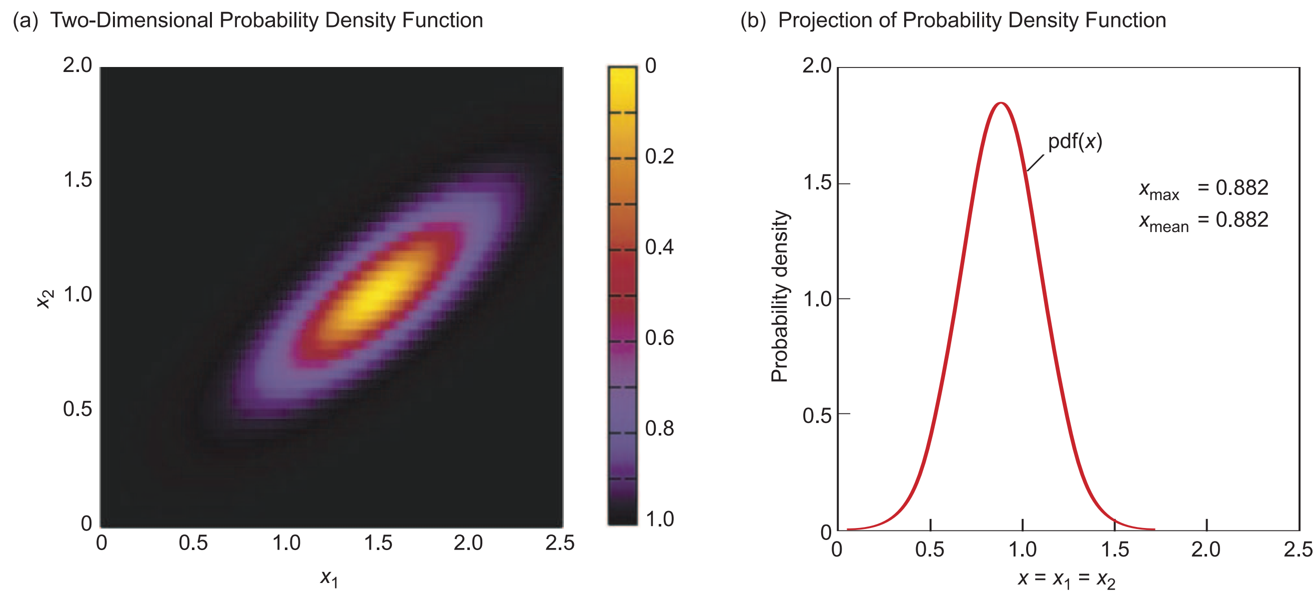


Figure 4. Peelle's Pertinent Puzzle (PPP)
Robert Peelle introduced the puzzle that now bears his name to illustrate the importance of including systematic errors in nuclear data evaluations. In his original example, there are two measurements of the same physical quantity, and the results are 1.5 and 1.0 respectively. Each result has a 10 percent uncertainty, and both results share a 20 percent common error. Standard statistical tools applied to this case give a best-estimate value of 0.882 for the physical quantity, which falls below both measured values! In (a), the two-dimensional Gaussian probability distribution function for the two measurements pdf(x_1,x_2) is shown; in (b), the projection of this distribution is shown on the $x_1 = x_2$ line, with the mean and maximum values equal to 0.882. However, this value depends on an underlying assumption regarding the nature of the correlated uncertainty. In practice, this knowledge is not often available.

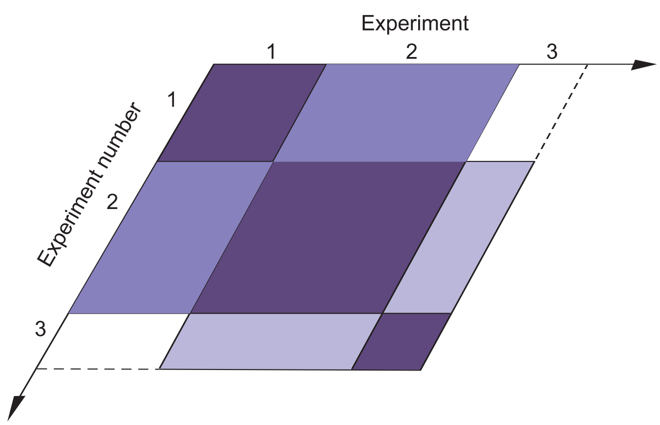
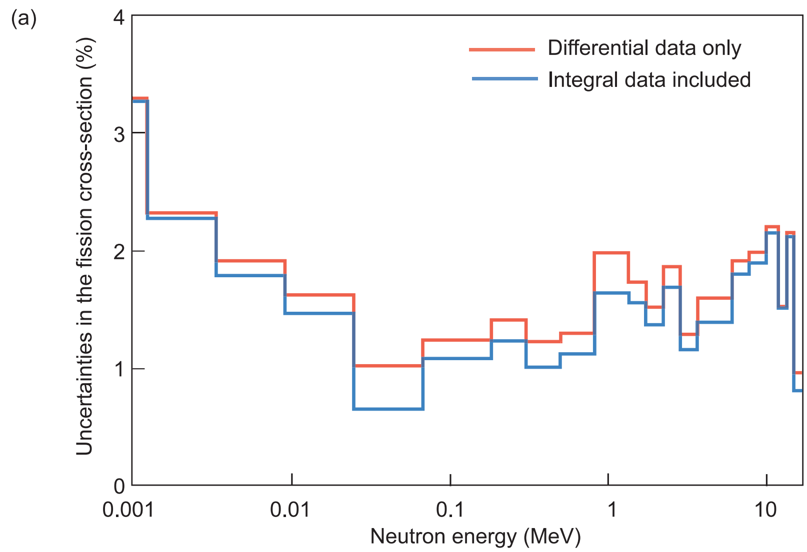


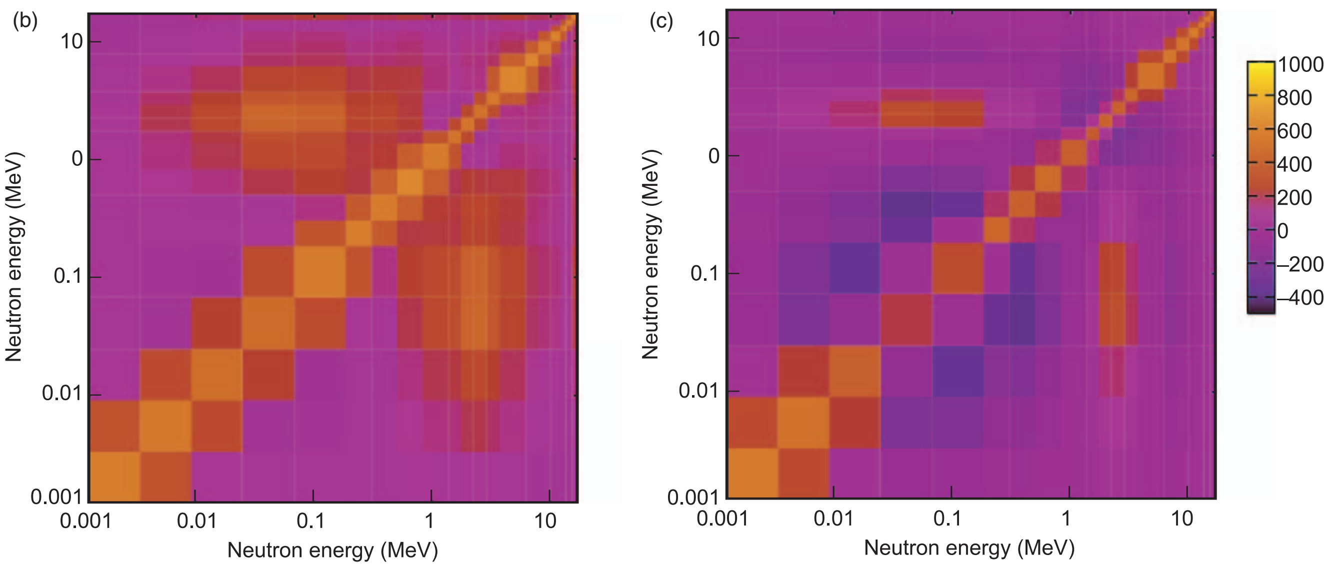
Figure 5. Representation of a Covariance Matrix
The role of a nuclear data evaluator includes constructing covariance matrices that completely describe the experimental data sets and the associated uncertainties and correlations for a given nuclear cross section. Each experimental set corresponds to an ensemble of cross-section values for various incident neutron energies. Statistical uncertainties are commonly given, whereas sources (and quantification) of systematic errors are only sometimes available. Correlations between different data sets can also exist—for example, if the same experiment facility, detector, or sample target is used in two distinct experiments. This picture shows a schematic representation of a corner of the large covariance matrix that results from the study of all the cross section data at all energies.

analysis. Unfortunately, they are also the most challenging quantities to estimate.

A fascinating example of the role of correlations in statistical analysis is Peelle's Pertinent Puzzle, or PPP for short (refer to Figure 4) named after Robert Peelle, who confronted the nuclear data community with a counterintuitive example. Suppose that two measurements of the same physical quantity are made, and the results are 1.5 and 1.0 respectively. Each result has a 10 percent uncertainty, and both results share a 20 percent common error. Standard statistical tools applied to this case give a best-estimate value of 0.882 for the physical quantity, which falls below both measured values! This result may be correct depending on the nature of the correlated uncertainty, additive or multiplicative. Because in many instances

Figure 6. Evaluated Variance-Covariance Matrix for the Pu-239 Fission Cross Section
In (a), the evaluated variances corresponding to the evaluated Pu-239 fission cross section (shown in Figure 3) are given when only differential data are used (red) and when integral data (blue) from critical assembly experiments are also included in the analysis. The corresponding correlation matrices are shown in (b) and (c), before and after inclusion of integral data in the analysis, respectively. The impact of adding integral information into our statistical analysis is clearly seen: It tends to reduce the standard deviations and generate negative correlation values that strongly constrain the fission cross section.





we do not know the origin of uncertainties, PPP represents a real puzzle for nuclear data evaluators, confronted with older and not well-documented experimental data.

The result of our comprehensive statistical analysis is depicted in Figure 3, along with the experimental data sets. The representation of the uncertainty with simple error bars on individual points is only part of the story. The covariance matrix for the evaluated cross section is also quite important. Figure 5 shows a schematic view of a portion of a covariance matrix that represents uncertainties and correlations among all experimental data sets included in the statistical analysis. The actual covariance matrix for the evaluated fission cross section is shown in Figure 6.

In summary, the correct estimation of experimental uncertainties and correlations is undoubtedly the most important aspect of precisely evaluating the plutonium fission cross section (and its uncertainties) in this kind of statistical analysis. Our project has benefited from extensive expertise by scientists at many institutions—especially at Los Alamos, the National Institute of Standards and Technology, and at the International Atomic Energy Agency—that have a long history of understanding and assessing the uncertainties and correlations in previous cross-section measurements.

Critical-Assembly Constraints on Fission Cross-Section Data. We have discussed how uncertainties on the plutonium fission cross sections can be determined from a Bayesian analysis of the experimental cross-section data. Next we show how integral measurements of the critical mass of plutonium are allowing us to make much larger reductions in uncertainty. Our ability to accurately model a critical assembly of plutonium using the MCNP transport code in conjunction with our neutron cross section data provides constraints on the uncertainties on the underlying microscopic plutonium fission cross-section data.

MCNP was developed at Los Alamos over many decades and is the world's most widely used, sophisticated, and well-tested code for simulating the coupled transport of neutrons and photons as they interact with nuclei. The interactions of neutrons with individual nuclei are modeled using nuclear cross sections from the evaluated neutron data files (ENDF) database developed at Los Alamos and other national laboratories. The accuracy of the transport calculational methods is so high that MCNP simulations of integral experiments, such as the criticality of a sphere of plutonium, provide a valid test of the accuracy of the underlying ENDF nuclear cross sections such as neutron-induced fission.

The calculated critical mass of plutonium depends on cross sections for a number of different neutron-plutonium interactions. It depends on the neutron-induced plutonium fission cross section, the average number of prompt neutrons ($\bar{\nu}$) emitted from fission fragments after a plutonium nucleus fissions, the cross sections for inelastic scattering of neutrons by plutonium nuclei; the angular distributions of neutrons that scatter elastically from a plutonium nucleus; and the cross section for a plutonium nucleus to capture a neutron. Of these quantities, it is the first two, and more precisely the product of the fission cross section and $\bar{\nu}$, that most sensitively influence the calculated critical mass and the neutron multiplication rate k_{eff} in the system, which equals unity when the system is critical.

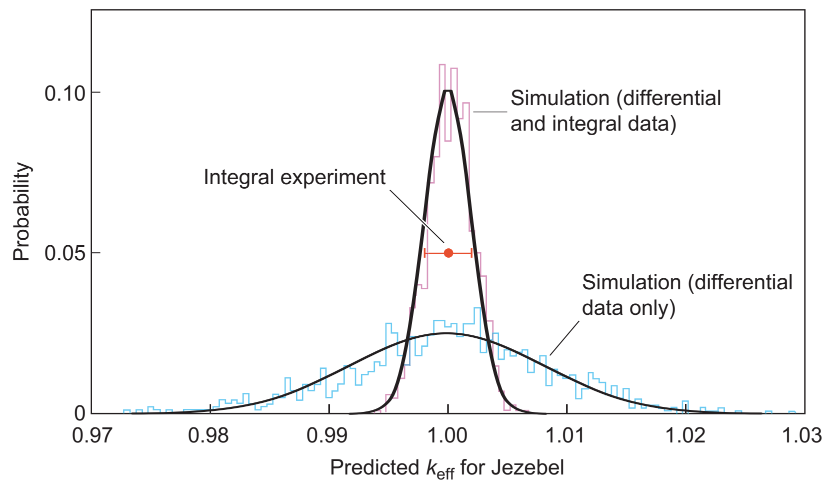


Figure 7. Probability Distribution Function for Jezebel's Neutron Multiplication Rate

We used our Bayesian uncertainty quantification code KALMAN to combine the prior information on differential fissions cross section measurements with the integral information from the Jezebel critical assembly measurement. The analysis provides posterior fission cross sections for different neutron energies. The simulation of Jezebel's criticality using those posterior cross sections yielded a probability distribution for k_{eff} (pink curve) that has a much smaller variance than that of our initial result from differential data only (blue curve).

If we were to estimate the fission cross section and $\bar{\nu}$ uncertainties based on only the fundamental, measured differential cross-section data discussed in the previous section, we would obtain uncertainties in the range of 1 to 2 percent for the fission cross section and less than 1 percent for $\bar{\nu}$, for neutrons with energies in the fission-spectrum energy range of 1 to 2 MeV. In an MCNP transport simulation of Jezebel, these numbers would translate into calculated uncertainties in the range of 1 to 2 percent for calculated values of k_{eff}.

However, Jezebel's measured criticality defines the k_{eff} uncertainty to less than 0.2 percent—an order of magnitude smaller than our previously calculated results based on cross section and $\bar{\nu}$ data uncertainties. We have used those integral measurements which are simple and highly accurate to constrain the differential fission cross sections by using the standard Bayesian technique. With this method, we were able to reduce uncertainties in the fission cross section, and the combined differential and integral data now predict that the neutron multiplication due to fission (k_{eff}) is accurate to about 0.2 percent, an order of magnitude more precise.

The plots in Figure 6 illustrate the uncertainty reductions. The uncertainties (variance and covariance) associated with the statistical analysis of the differential experimental data alone are shown (red line) in Figure 6(a) (the variance) and in Figure 6(b) (the correlation matrix multiplied by 1000). Neutron transport calculations were performed for the Jezebel critical assembly, and the sensitivity coefficients of the cross sections to the neutron multiplicity were obtained. Then Jezebel data were used to adjust the fission cross section through the Bayesian inference method. The resulting uncertainties in the fission cross section are shown in Figure 6(a) (blue line). The impact on the fission cross section itself is very small. However, the uncertainties become smaller, and negative correlations appear, as shown in Figure 6(c).

These negative correlations constrain the fission cross sections in order to keep the integral quantities constant. If we generate randomly sampled fission cross-section ensembles in accordance with this covariance matrix, the calculated values of k_{eff} for Jezebel form a Gaussian distribution of 0.2 percent uncertainty. This result can be seen in Figure 7, where the large reduction in the uncertainty of the calculated criticality is evident by comparison with the uncertainty from methods that do not use integral measurements.

Iridium Nuclear Cross Sections

Nuclear weapons performance is affected by the neutrons the weapons produce. The neutrons induce nuclear fission in the plutonium and uranium components of the device, and a runaway fission chain reaction occurs that releases the massive amount of energy driving the nuclear explosion. Many of the variables that affect weapons performance depend on the energy distribution (spectrum) of the neutrons. The neutron energy spectrum, for example, determines the relative rate at which fission occurs versus other neutron-induced nuclear reactions, and it also determines the number of neutrons released per atom during the fission process.

Certain elements have been used almost since the inception of the nuclear age to gain spectral information about the all-important neutrons. Small amounts of these so-called radiochemical (radchem) detector materials were placed in specific locations within a nuclear weapon before a test. During the explosion, the intense neutron flux transmuted some of the atoms of the detector material into other, predominantly radioactive, isotopes. After obtaining tiny amounts of the postshot test debris, radiochemists would extract the detector element from the samples and measure the relative amount of each radioactive isotope. Provided the nuclear cross sections for the production and/or destruction of the stable and radioactive isotopes were well understood and measured accurately, a weapons designer could relate the isotopic ratios to the neutron fluence[4] that occurred within the device.

During the era of nuclear weapon testing, different radchem detectors were used to measure the neutron fluence in different energy ranges. Certain nuclear reactions—for example, the $(n,2n)$ reaction, in which one neutron impinges on a nucleus and two neutrons are emitted—are known as threshold reactions; they occur only if the energy of the incident neutron is above some threshold energy, typically a few million electron volts or higher. Isotopes that are produced by the $(n,2n)$ reaction were used to measure the high-energy (about 14 MeV) neutron fluence produced by fusion reactions. Other reactions for producing new isotopes, notably the (n,γ) neutron capture process (in which a nucleus captures an incident neutron and emits a gamma ray), have no threshold. Neutron capture is more likely to occur as the neutron energy decreases and (n,γ) neutron capture reactions dominate isotope production when the neutron energy is below 1 MeV.

The reaction that has been used as a diagnostic for neutron energies between these two extremes is the (n,n') inelastic neutron-scattering reaction in which an iridium-193 nucleus absorbs some energy from the incident neutron and transitions to a long-lived nuclear excited state known as the isomer iridium-193m. This reaction is uniquely sensitive to neutrons with energies in the few-million-electron-volt range, which, in turn, is the energy range of neutrons produced in the fast chain reaction in a weapon. Hence, determining the production of the isomer iridium-193m is an extremely important diagnostic for weapon performance.

Figure 8 indicates with arrows the reaction pathways that can occur when neutrons are incident on an iridium target composed of the stable isotopes 191 and 193. By measuring the production of radioactive iridium-189, -190, -192, 193m, and –194 in such a target, one can learn information about all three energy-sensitive neutron-induced reactions, $(n,2n)$, (n,n'), and (n,γ). Iridium, therefore, provides a unique diagnostic capability of the neutron fluence in multiple energy regimes, including the few-million-electron-volt fission neutron-energy region.

Unfortunately, measuring the amount of iridium-193m produced in a nuclear test was also uniquely difficult. It must be done by measuring the decay of the radioactive isomer, but the decay proceeds through two competing processes, gamma-ray emission and internal conversion, and the latter is very difficult to separate from the background[5]. The problem was first solved by some of the great figures from the radchem past of Los Alamos, such as Jim Gilmore, Don Barr, and Moses Attrep. The experimental problem was so difficult that other laboratories, such as Lawrence Livermore

[4] Radiochemistry measures only time-integrated quantities because its measurements reveal the cumulative result of a long, complex sequence of production-destruction reactions on the nuclei.

[5] Internal conversion is a nuclear decay process in which the nucleus changes to a lower energy level and maintains energy conservation by emitting an electron from an atomic shell. Because it is charged, that electron tends to be stopped in the sample, emitting x-rays as it slows down. Often those x-rays can be very difficult to separate from the background. In the more usual decay process, the nucleus emits a readily detected gamma ray as it decays to a lower energy level.

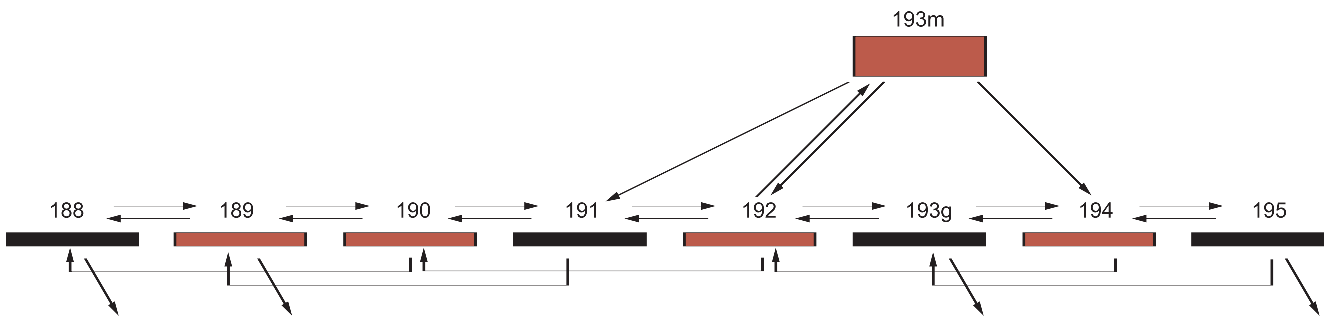


Figure 8. Reaction Pathways for Neutrons Hitting an Iridium Target

The different arrows correspond to neutron-induced reactions on iridium nuclei such as (*n*,), (*n*,*n′*), (*n*,2*n*) and (*n*,3*n*), where the left entry indicates the incident particle and the right entry indicates the outgoing particles. By measuring the various production rates of the radioactive isotopes iridium-189, -190, 192, -193m, and -194 when exposed to a particular neutron fluence, one can learn precious information on the cross sections for each reaction present in this reaction network. In particular, the inelastic neutron scattering reaction cross section for iridium-193 (*n*,*n′*) iridium-193m reaction cross section is most sensitive to neutrons in the few-million-electron-volt energy range and therefore can contribute to assessing the neutron fluence in this neutron energy range.

National Laboratory (Lawrence Livermore) and the Atomic Weapons Establishment in Great Britain, relied upon Los Alamos radiochemistry for this task.

Nuclear Cross Sections and Uncertainty Quantification. As mentioned earlier, to accurately infer neutron fluences from radiochemical measurements of isotopes after a nuclear test, it is not enough to determine the relative amounts of the various isotopes. The nuclear cross sections for producing those isotopes must also be known accurately. This has not been the case for iridium cross sections. In particular, the (*n*, *n′*) neutron-scattering cross section that determines the production of the isomer iridium-193m is extremely difficult to measure because there are many different pathways leading to isomer production and some of them—for example, direct population of the isomer state through neutron scattering and internal conversion—cannot be observed. Figure 9 shows a diagram of the energy levels of the iridium nucleus and the many pathways that lead to population of the isomeric state.

Until recently, the only experimental data on iridium isomer production were obtained at incident neutron energies above 7.5 MeV by the Los Alamos radchem group mentioned (Bayhurst et al. 1975). Consequently, the historic isomer production cross-section data set used at Los Alamos for the last two decades was based almost exclusively on the nuclear-theory predictions of Ed Arthur of the Theoretical (T) Division at Los Alamos.

In the last few years, a collaboration between experimentalists at the Los Alamos Nuclear Science Center (LANSCE) and theoreticians in T-Division has determined and evaluated new data for the isomer-production cross section. LANSCE's GEANIE gamma-ray detector (see Figure 10) was used to measure the cascade of gamma rays that results when the excited iridium-193 nucleus loses energy on its way to populating the metastable isomeric state. The GEANIE measurements were undertaken by a Los Alamos–Lawrence Livermore collaboration involving Ron Nelson, Nick Fotiadis, Matt Devlin, John Becker, Paul Garrett, and Lee Bernstein. But GEANIE could not measure the contributions to the isomer production from processes that do not involve gamma rays. Those contributions had to be predicted from theory. Theory was also needed to predict certain gamma-ray feeding transitions that could not be measured directly because of experimental limitations.

The authors accomplished that task by incorporating advanced nuclear-reaction-theory models into the GNASH code, which was developed in T-Division for predicting nuclear cross sections. Those advanced models describe compound nucleus, pre-equilibrium, and direct mechanisms for a nucleus to reach an isomeric state. In order to accurately model isomer production, we had to understand the following nuclear properties: (1) optical potentials that describe the motion of the incoming and outgoing neutrons relative to the target nuclei, (2) the nuclear structure and decay properties of the low-lying levels (obtained from experiment) and highly excited levels (obtained from statistical theories of excited nuclei), and (3) the angular momentum transfer processes associated with the pre-equilibrium and compound nucleus decay mechanisms. Our

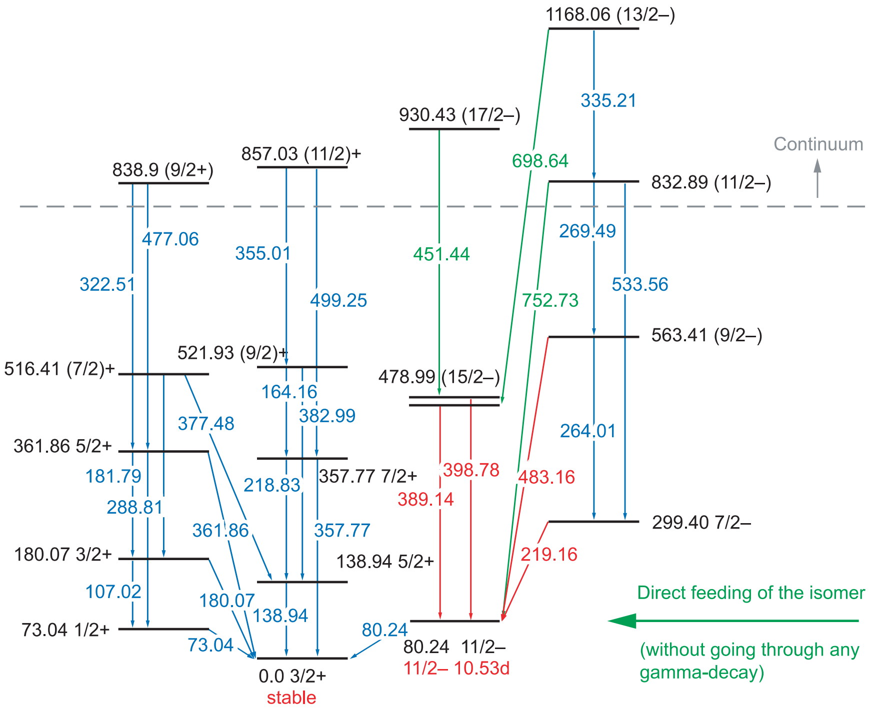


Figure 9. Pathways for Producing the Isomer Iridium-193m

This nuclear energy-level diagram shows the various pathways for producing the long-lived isomer state at an energy of 80 keV above the ground state. The GEANIE experiment clearly resolved the four strongest γ-ray transitions (red lines) that feed the 80-keV isomer. GNASH calculations were benchmarked against the GEANIE data for the strengths of those four transitions and then were used to calculate all other unaccounted for contributions to the isomer production cross section. The latter include the direct feeding of the isomer by neutron inelastic scattering (without going through the γ-ray cascade) and the other γ-ray transitions (green lines) that either reach the isomer or feed levels that reach the isomer.

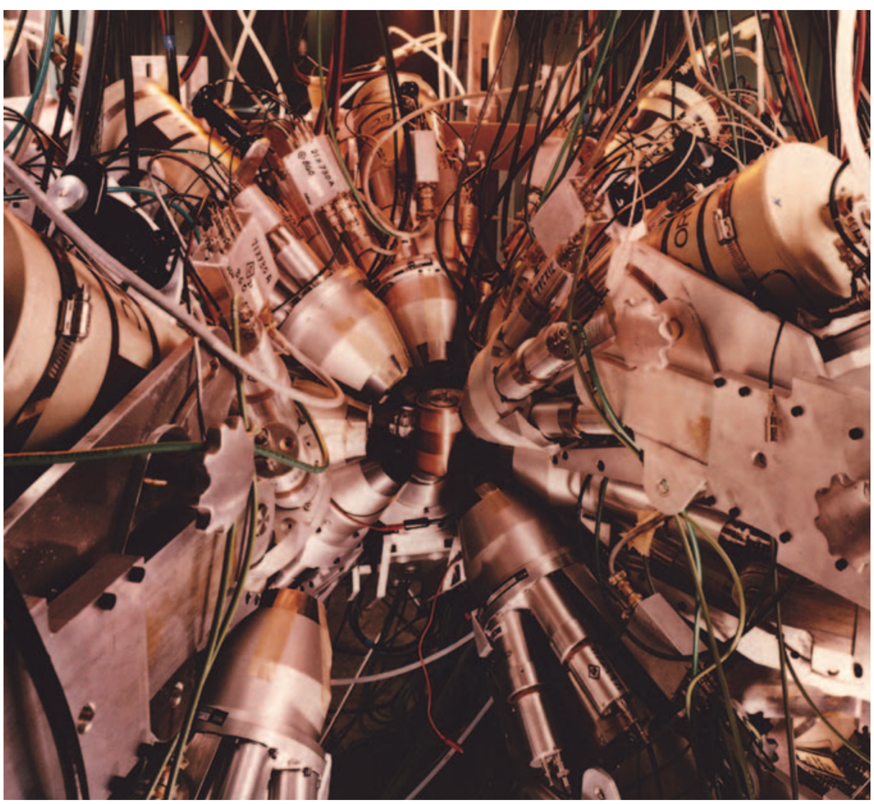


Figure 10. The GEANIE Detector

GEANIE (germanium array for neutron-induced excitations) is a 4π high-resolution γ-ray spectrometer installed at LANSCE's Weapons Neutron Research Facility. It can detect γ-rays from about 20 keV up to 8 MeV. The neutrons hitting the target samples cover the energy range from below 1 MeV to more than 200 MeV. The time-of-flight technique is used to determine precisely the energy of the incident neutrons, with a 22-m flight path. The GEANIE spectrometer was used to study details of the γ-ray cascade following the inelastic neutron scattering on iridium-193.

Figure 11. New Evaluated Production Cross Section for Iridium-193m
The new GEANIE/GNASH prediction for the 80-keV isomer production cross section in iridium-193 is shown here, covering the incident neutron energy range from the reaction threshold (80 keV) up to 20 MeV. The 1-σ standard deviations that come from uncertainties in both GEANIE data and GNASH reaction modeling are also plotted. This new cross section is compared with the historic one from T-Division (by Ed Arthur and Robert Little) that has been used until now in weapon physics work at Los Alamos. Note that our new result is in much better agreement with the MacInnes ad hoc fix to the Arthur-Little evaluation near threshold, which was incorporated to improve the agreement with data from critical assemblies.

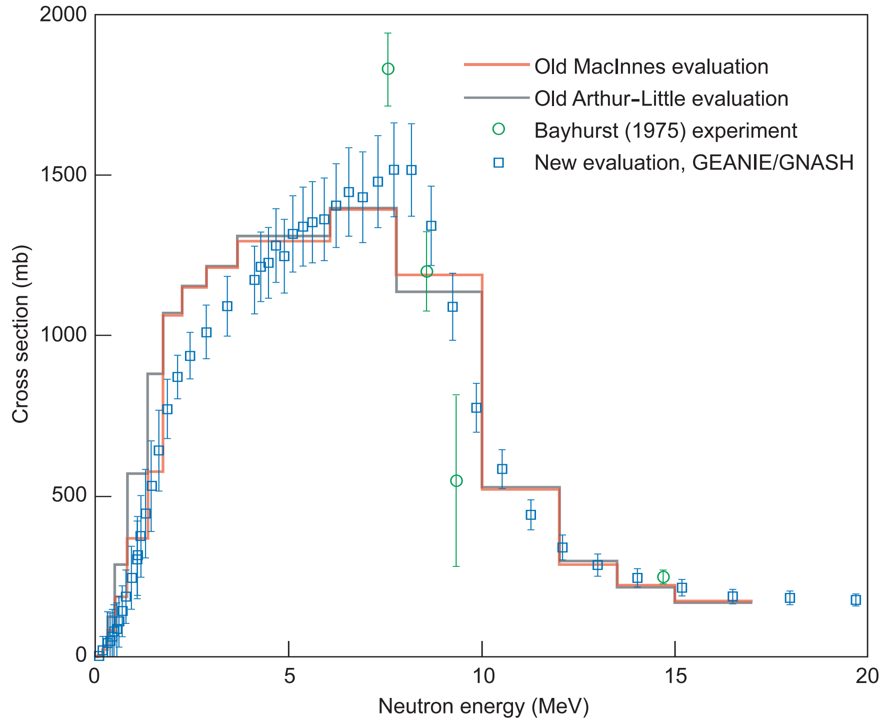
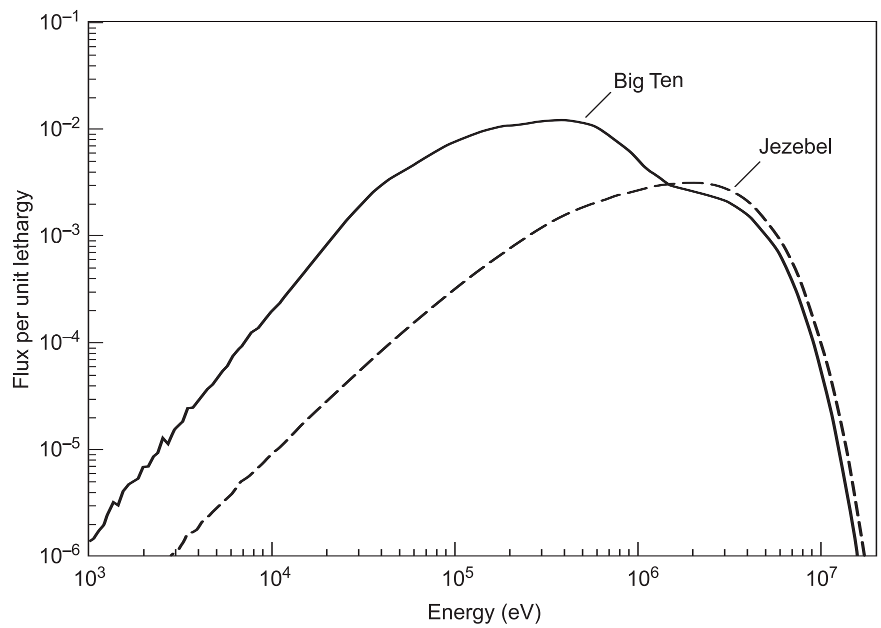


Figure 12. Hard and Soft Neutron Spectra
Assemblies with the highest average neutron energy are said to have the "hardest" spectra, whereas those that produce neutrons with a lower mean energy are said to have "softer" spectra. For example, the center of the Jezebel assembly (a sphere of plutonium) has one of the hardest spectra available. The Big Ten assembly, which has large amounts of uranium-238 and -235, has a much softer neutron spectrum.



research on these properties for iridium, together with extensive experience we have built up in analyzing similar data for other nuclei measured at LANSCE, allowed us to predict the various contributions to iridium isomer production using our advanced version of the GNASH code.

To test the accuracy of our calculational ability, we compared our GNASH cross-section predictions for the measured gamma-ray decay transitions with those determined from the GEANIE measurements. After we validated our predictive capability, we could apply the theory to predicting the unmeasured contributions with confidence. We could then evaluate the isomer-production cross section and its uncertainty using both the GEANIE and GNASH results.

Figure 11 shows our newly evaluated cross section for isomer production. The new GEANIE-GNASH

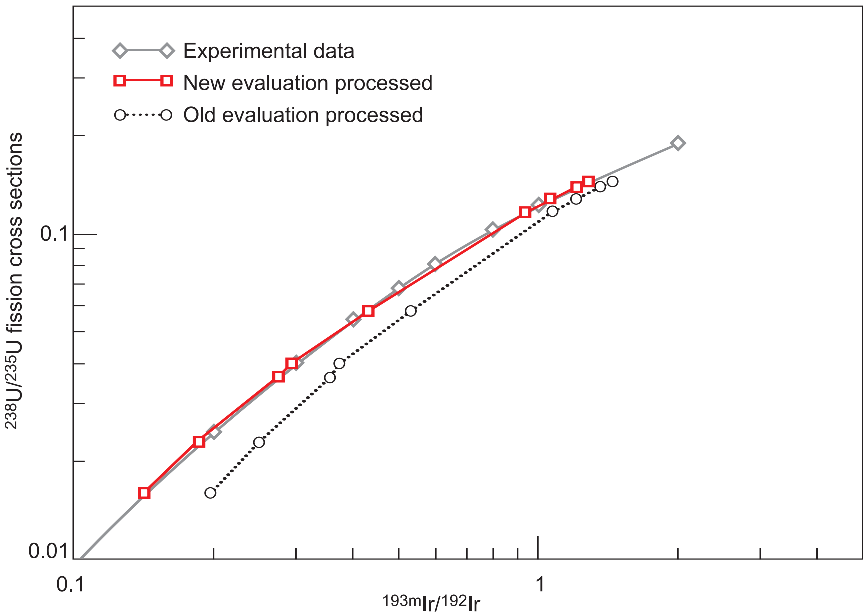


Figure 13. Iridium-193m Production Cross Section

Experimental data obtained with critical assemblies at LACEF were used to validate our new evaluation work. This figure represents the ratio of the iridium-193m production cross section to the production of iridium-192 (mainly through the neutron capture cross section of iridium-191) as a function of the ratio of uranium-238 to uranium-235 neutron-induced fission cross sections. The latter ratio represents the "hardness" of the neutron spectrum. This quantity changes with the location of the target in the critical assembly. Near the center, the neutron spectrum is quite hard; at larger distances, it softens. The slope of the experimental curve in this figure is therefore an indicator of the shape of isomer production cross-section, in particular near the threshold energy. Our new evaluation represents a net improvement over the older existing evaluation.

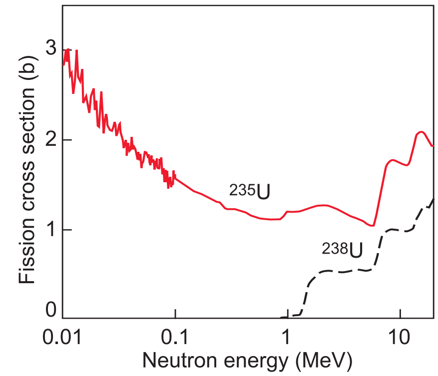


Figure 14. Fission Cross Sections for Uranium-235 and Uranium-238

Note the rapid increase of the uranium-238 fission cross section around an incident neutron energy of about 1 MeV. This threshold behavior causes the ratio of the uranium-238 to uranium-235 cross sections in critical-assembly experiments to increase with spectral hardness.

results cover the whole energy range of interest, from the threshold of the reaction at 80 kilo-electron-volts (keV) to above 20 MeV. Ed Arthur's old theoretical evaluation is also shown. Although the two results are similar overall, they also differ in subtle but important ways. In particular, our new cross section rises from threshold in a different manner, with a steeper slope. This outcome has important consequences, as will be described in more detail below. The uncertainties that we have derived for this cross section are shown as 1-σ error bars in Figure 11. The uncertainties that we have deduced include systematic and statistical errors, and they are associated with both the measured data and the GNASH nuclear model calculations.

Integral Data Testing at Critical Assemblies. With our new cross sections in hand, we will undertake weapon code simulations of specific past underground nuclear tests in which the nuclear devices were loaded with iridium radchem detectors and combine the calculated neutron fluences and our new cross sections to predict the iridium isotopic ratios produced in those tests. Because we have determined cross section uncertainties for the iridium reactions, we will also be able to provide uncertainties on the weapons code predictions of the iridium isotopic ratios. We will then compare the predicted ratios against actual post-test radchem measurements from those tests. Finally, we will work with designers to incorporate the results of those comparisons into the baseline certification calculations.

Interestingly, we have been able to validate our new iridium cross sections against an old but fascinating and unclassified set of iridium radchem data. Those data, obtained from fast critical-assembly experiments conducted over several decades at LACEF, provide a valuable integral test of our iridium cross sections. The fast critical assemblies at Los Alamos involve macroscopic quantities of special nuclear materials—plutonium and uranium-235 and -238—often in spherical configurations. When the critical mass is assembled, a self-sustaining chain reaction occurs, creating a neutron flux that has the energy spectrum typical of a fast fission-chain reaction. During the iridium radchem experiments, the flux of neutrons irradiated iridium foils placed inside the assembly, and the ratios of various iridium isotopes produced during irradiation were subsequently measured. One such ratio was iridium-193m/iridium-192, in which the isomer came from the iridium-193

(n,n') reaction and the isotope, from the iridium-191 (n,γ) reaction. (Contributions from the iridium-193 (n,2n) reaction are very small in a critical assembly.)

Those old measured ratios can be compared with new predictions for these ratios obtained with our new cross sections. To predict the isotopic ratios, we must first predict the neutron energy spectrum of the fast critical assembly experiments using an MCNP radiation transport simulation and then fold that spectrum together with our iridium cross sections.

Clearly, fast critical assemblies provide valuable integral experiments to test our iridium cross sections because the neutron energy spectrum created in a fast critical assembly is skewed toward neutron energies that the iridium-193m diagnostic was developed to detect, that is, energies in the few-million-electron-volt region. But we also wanted to validate our cross sections over energies extending down to the threshold for isomer production, which is 80 keV. Again, iridium radchem data from old critical-assembly experiments have been invaluable. The various assemblies provide neutron energy spectra with varying average energies, depending on the critical assembly and the location within that assembly (see Figure 12). Fortunately for our iridium work, radiochemists had already conducted experiments in which iridium foils were loaded at various radial locations throughout a "traverse" of the Flattop assemblies (a core of HEU or plutonium surrounded by uranium-238). Those experiments involved neutron spectra ranging from "hard" (at the center of the assembly) to "soft" (at maximum distance from the center).

Figure 13 shows the radiochemical results obtained with the Flattop assembly for the isotopic ratio of iridium-193m to iridium-192 as a function of the hardness of the critical-assembly neutron spectra, where the spectral hardness is represented by the ratio of uranium-238 to uranium-235 fission cross sections. That fission cross-section ratio is used for two reasons: It increases with spectral hardness, or average neutron energy (see Figure 14), and it can be measured within the assembly, at the very spot where the iridium foils have been placed. Figure 13 also shows our calculated results for the iridium isotopic ratios, as well as results from Ed Arthur's old evaluation. The good agreement between measured data and our new calculated results validates our iridium-193m (n,n') cross section in the few-million-electron-volt region. Moreover, our reproduction of the shape of the experimental curve derived from the Flattop integral experiments validates the shape of the new GEANIE/GNASH microscopic cross section in Figure 11 as it rises from threshold.

The validation of the new isomer production cross section near threshold represents a breakthrough. Several years ago, Mike MacInnes of Los Alamos first undertook calculations of the Flattop critical assembly data in Figure 13 with the historic Ed Arthur's iridium-193 (n,n') isomer cross section used at Los Alamos at the time. He noted that the calculated shape did not agree well with the measured shape. This observation led him to make a change to the shape of the historic cross section near threshold. Our new result for this same cross section, based on independent LANSCE data and nuclear model calculations, has confirmed MacInnes' intuition.

Conclusions

Our ability to predict important nuclear cross sections and quantify uncertainties in those predictions has advanced considerably in the last decade. The rates of neutron-induced fission reactions are crucial to the performance of weapons. That is why reducing the uncertainty in those rates leads to more confident predictions using the Los Alamos weapons simulation codes. In addition, increased accuracy of neutron-scattering results obtained with radchem tracers has contributed to better assessments of past nuclear tests. ■

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