INEEL/EXT-03-00372

Nuclear Data Measurements for 21st Century Reactor Physics Applications

Technical Basis Document

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Idaho National Engineering and Environmental Laboratory
Bechtel BWXT Idaho, LLC

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CONTENTS

ACRONYMS

AFC	Advanced Fuel Cycle
ANL	Argonne National Laboratory
CSHPGe	Compton–suppressed, high-purity germanium
CSWEG	Cross Section Evaluation Working Group
DOE	Department of Energy
EM	Office of Environmental Management
ENDF	Evaluated Nuclear Data File
ESRA	Environmental Science Research and Applications
FY	Fiscal Year
Gen-IV	Generation-IV
INEEL	Idaho National Engineering and Environmental Laboratory
IPNS	Intense Pulsed Neutron Source
LANL	Los Alamos National Laboratory
LANSCE	Los Alamos Neutron Science Center
LLNL	Lawrence Livermore National Laboratory
LWR	Light Water Reactor
MTR	Materials Test Reactor
NE	Office of Nuclear Energy
NERAC	DOE Nuclear Energy Research Advisory Committee
NDAG	DOE Nuclear Data Advisory Group
NNDC	National Nuclear Data Center
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PBF	Power Burst Facility
PSO	Program Secretarial Office
R&D	Research & Development
SNS	Spallation Neutron Source
WNR	Weapons Neutron Research

SUMMARY

The United States Department of Energy (DOE), Office of Nuclear Energy (NE) has embarked on a long-term program to significantly advance the science and technology of nuclear energy. This is in response to the overall national plan for accelerated development of domestic energy resources on several fronts, punctuated by recent dramatic events that have emphasized the need for the US to reduce its dependence on foreign petroleum supplies. Key aspects of the DOE-NE agenda are embodied in the Generation-IV (Gen-IV) advanced nuclear energy systems development program and in the Advanced Fuel Cycle (AFC) program. The planned efforts involve near-term and intermediate-term improvements in fuel utilization and recycling in current nuclear power reactor systems as well as the longer-term development of new nuclear energy systems that offer much improved fuel utilization and proliferation resistance, along with continued advances in operational safety.

The success of the overall NE effort will depend not only on sophisticated system development and engineering, but also on the advances in the supporting sciences and technologies. Of these, one of the most important is the improvement of the relevant fundamental nuclear science data bases, especially the evaluated neutron interaction cross section files that serve as the foundation of all reactor system designs, operating strategies, and fuel cycle engineering activities. The new concepts for reactors and fuel cycles involve the use of transuranic nuclides that were previously of little interest, and where experimentally measured information is lacking. The current state of the cross section database for some of these nuclides is such that design computations for advanced fast-spectrum reactor systems and fuel cycles that incorporate such materials in significant quantities are meaningful only for approximate conceptual applications. No actual system could reliably be designed according to currently accepted standards, nor could such a system be safely and efficiently operated, with the limited nuclear data and related information now available.

In order for the required new information to be available, with the appropriate level of validation and evaluation of quality, in the 10-20 year time frame envisioned for development of the various advanced reactor and fuel cycle concepts, it is necessary to undertake a concerted effort focused on this objective in the very near term. Technologies and capabilities within the US National Laboratory system and collaborating universities and international institutions are available to address the nuclear data issue, and some limited preliminary efforts are already underway. However, the lead times for this type of supporting scientific research are very long and the necessary measurements are exacting and complex.

The Idaho National Engineering and Environmental Laboratory (INEEL) is in an excellent position to support needed improvements in relevant fundamental nuclear science databases through a collaborative directed measurements program based on the most current capabilities present in the low-energy nuclear physics community. A team from the INEEL, in collaboration with Argonne National Laboratory (ANL), has been involved in an internationally recognized measurement program studying the prompt fission process and important results have been obtained and extensively published over the past 10 years. This program is built upon the advances in nuclear measurement instrumentation and data processing techniques that offer a new approach for studying the fission process with unprecedented improvements in data quality and resolution. The same basic experimental techniques, which involve accelerator-based pulsed neutron systems and sophisticated time of flight spectral resolution, coupled with advanced electronics and computational methods to collect and analyze the interaction data, can also be used for high-precision cross section measurements of interest to DOE-NE.

Some initial relevant studies are also underway elsewhere. In particular, Los Alamos National Laboratory (LANL) has recommended the performance of some initial actinide cross section measurements at the Los Alamos Neutron Science Center (LANSCE) in the most recent (Rev C, November 20, 2002) draft of the AFC Program Plan. These measurements should provide some useful preliminary data, at least for some neutron capture interactions over part of the energy range of interest. In addition to the fission studies work at ANL, members of the INEEL team have historically also collaborated with LANL in connection with various nuclear data measurements of interest to the environmental remediation community, and this team is well-poised to continue this collaboration for the new applications required for the NE mission. Some additional cross section measurements for non-transuranic nuclides have been underway at the Oak Ridge ORELA facility. These are primarily focused on the needs of the criticality safety and astrophysics communities, but some of the results would also be of interest to the reactor physics community. On the international front, there are plans for various types of relevant cross section measurements at the CERN n-TOF facility (Tassan-Got, 2001), although this effort has encountered some recent serious technical challenges (Cano-Ott, 2001).

The INEEL, in a continued collaboration with ANL, can expand and redirect its current fission research program in a manner that will contribute new and useful information relevant to the actinide cross section issue in the very near term. A multi-element array of Compton–suppressed, high-purity germanium (CSHPGe) detectors, operated in coincidence mode, with sophisticated data collection and analysis techniques has been installed by the INEEL at the ANL/Intense Pulsed Neutron Source (IPNS) facility and is now in operation. Data for all observable neutron interactions occurring in the sample target are collected simultaneously during one experimental cycle and detailed information for the interactions of specific interest is extracted later, all from the same data set, eliminating the need to run separate experiments for each interaction. Because of this feature, the INEEL system at ANL/IPNS, with some straightforward modifications, can be used to collect high accuracy cross-section data for many of the exotic actinide species of interest to Gen-IV and AFC, as noted previously. For example, some initial measurements for Np-237, one of the highest-priority nuclides for which improved data are needed can begin within the next 2 years at ANL/IPNS, using a multi-gram sample target already available at ANL/IPNS. This capability will also be very useful for independent validation of cross section measurements performed at other facilities - a key component in the accepted data evaluation process. Additional targets for other nuclides of interest can be obtained as needed from well-established sources in Russia.

In addition to the immediate capabilities of the INEEL setup at ANL/IPNS, several key improvements can be made in the near future to produce even more useful and higher quality results while simultaneously reducing the time required for a given measurement. The experimental apparatus can be upgraded with additional detectors and electronics to further improve the signal to noise ratio and to dramatically (by a factor ~10) reduce the time required to achieve the desired level of statistical accuracy in the results. In the intermediate term, a similar system could be installed on a more intense dedicated beam line at IPNS to provide significantly improved quality at modest additional expense. In the longer term, some work could also possibly be done at the Oak Ridge National Laboratory (ORNL)/Spallation Neutron Source (SNS) facility that is currently under construction at Oak Ridge. A dedicated beam line for this purpose would need to be developed by NE at ORNL/SNS, but the result could be an unparalleled facility for accurate, high resolution, measurement of nuclear data of interest to NE for many years to come.

Furthermore, over the longer term the new techniques described above can also be used to measure useful data for some non-fuel nuclides of interest to NE using much simpler and far more cost-effective neutron sources than the pulsed accelerator systems required for the more difficult measurements. A very simple steady-state neutron producing system using a Cf-252 source could be developed at the INEEL and would be useful for some measurements without tying up the heavily used and much more expensive accelerator systems that are currently available in the US. Such a system would also be useful as a facility for further development of the advanced custom electronics required for nuclear interaction studies, as well as for training of students in an environment that is free of the time pressures and scarce resource availability issues associated with the large, multi-user accelerator facilities.

The planned overall effort will compliment and substantially expand upon, not duplicate, other related efforts that are ongoing elsewhere, and multiple measurements with different methods will provide the necessary checks on all data.

1.0 INTRODUCTION

The Idaho National Engineering and Environmental Laboratory (INEEL) has a long history of contributions to the basic fields of nuclear science underlying the modern nuclear industry. The Materials Test Reactor (MTR) at the INEEL was used for many years to measure nuclear cross sections and other fundamental information. Many of the protocols and standards for modern gamma spectroscopy were also developed at the INEEL. Most recently, the INEEL has maintained a core capability in nuclear science under the Environmental Science Research and Applications (ESRA) Program, sponsored by the US Department of Energy (DOE) Office of Environmental Management (EM), which served for a number of years as the Program Secretarial Office (PSO) for the laboratory.

In July 2002 the INEEL experienced a major change in management direction, with the PSO being changed to the DOE Office of Nuclear Energy (NE), and the laboratory Research & Development (R&D) mission significantly reoriented to emphasize the needs of DOE-NE. This especially has impact in connection with the Generation-IV (Gen-IV) advanced reactor development program and the Advanced Fuel Cycle (AFC) Program, both operated by the division of Advanced Nuclear Research, NE-20. Furthermore, the INEEL was named by the Secretary of Energy as the focal point for nuclear energy research and development in the US, implying a significantly greater leadership role in this area than has been the case in the recent past. As a result of these new developments, key INEEL capabilities in nuclear science and engineering as well as in radiochemistry and several other fields will be directed toward the new mission to support a wide variety of nuclear energy related applications. Of specific importance here, our experience with performing various types of fundamental nuclear measurements to support DOE-EM is also directly applicable to similar activities of interest to DOE-NE, and will be refocused accordingly.

There is a recognized need for improvement of the neutron cross section data base for several of the actinides, especially in the case of nuclides other than U-235, U-238, and Pu-239, which of course already have been extensively studied and characterized. As discussed in the DOE-NE Long Term Nuclear Technology Research and Development Plan, published in June, 2000 by the DOE Nuclear Energy Research Advisory Committee (NERAC), there is a clear recognition in the U.S. community that, "*Advances in reactor concept design can be expected to require additional data for basic nuclear properties, such as neutron and gamma spectral data, microscopic cross sections (and other information). The existing data base is only marginally adequate for present applications and is unlikely to be sufficient for future applications*." A similar consensus exists in the international community (Nakagawa et al., 1999). The INEEL can make significant contributions to rectify this situation. In recent years, we have maintained unique capabilities to perform a variety of basic nuclear data measurements of interest for environmental applications. These capabilities can also be redirected to perform experimental measurements of the type required to improve and extend the neutron cross section database that is a key foundation for nuclear reactor design and nuclear fuel cycle analysis.

Figure 1 shows the so-called "minor actinide" buildup and decay chains that are most commonly considered in the design of advanced reactors and fuel cycles. These are of course in addition to the most fundamental chains involving U-235, U-238, and Pu-239. In future work the list may expand to include even heavier actinides (up to Cm-250 and possibly Bk-247). In most current nuclear power systems, which largely feature thermal neutron spectra, the minor actinide chains are of significance only because they lead to materials that must be managed with regard to the disposition of spent fuel from the "once-through" fuel cycles associated with these systems.

However, in more advanced systems, many of which will operate with fast neutron spectra, the minor actinides are viewed more as a resource to be recycled, or transmuted to less hazardous and possibly more useful forms, rather than simply as a waste to be disposed of in expensive repository facilities. As a result, they play a much larger part in the design of advanced systems and fuel cycles, not only as additional sources of useful energy, but also as direct contributors to the reactivity of the systems into which they are incorporated. However, as noted previously, the cross section database for these materials, which heretofore were not of much interest, is not currently adequate to perform the necessary design computations for the reactors and accelerator-based actinide transmutation systems that are contemplated under the Gen-IV and AFC Programs.

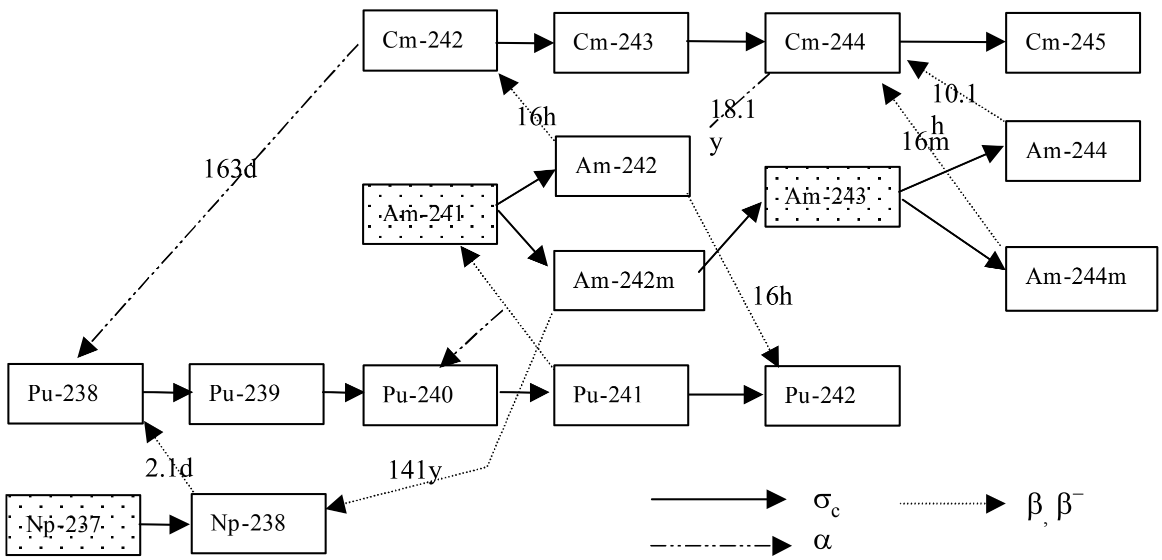

Figure 1. Buildup and decay chains for the minor actinides. Shaded boxes represent materials with long half-lives that make them of particular interest for transmutation.

In the following sections we provide a more detailed description of the current issues surrounding nuclear data needs for the minor actinides. This is followed by a summary of well-established INEEL technical capabilities that can, in the very near term, be brought to bear on the problem and a recommended path forward is offered.

2.0 BACKGROUND

The current status of the relevant cross section information for the minor actinides is summarized in this section. This is followed by a few examples showing how cross section uncertainties can propagate through various types of nuclear system design computations in a manner that can make the results nearly useless in some cases and highly subject to question in many other cases. In addition, a short description of current known US activities, outside of the INEEL, that is relevant to improving the situation is given.

2.1 Description and Status of the Evaluated Nuclear Data Files

Evaluated nuclear data (cross sections and other pertinent parameters) are required for computations and experimental support for a variety of applications including nuclear reactor physics, nuclear criticality safety, medical physics, radiation protection, and national security. The central repository for evaluated data in the US is the Evaluated Nuclear Data File (ENDF) maintained at National Nuclear Data Center at Brookhaven National Laboratory. The ENDF data archive, the data formats, processing procedures for the data, data testing activities (comparison of calculated results with integral measurements), and various documentation collections, are controlled and overseen by the Cross Section Evaluation Working Group (CSWEG), which is composed of representatives from the DOE laboratory system as well as other interested institutions.

In general, the priorities for measurement of neutron interactions and cross sections are determined by the scientific community based on fundamental research interests and in some cases by institutions or organizations, such as DOE, that have a particular and specific need. In the early history of nuclear research there was an overlap of scientific goals and national needs that drove the measurement programs but these formerly shared goals have diverged. In the current environment of large facilities and high cost of experimental equipment, only government-sponsored programs are now providing nuclear data.

The National Nuclear Data Center (NNDC) acts as a clearinghouse for collection and evaluation of information - they specifically do not manage or fund a formal integrated nuclear data measurement program. Nuclear data sets of interest to NNDC are measured by scientific investigators worldwide in connection with a variety of different national and international programs, and the results are published in the literature or otherwise made available. The ENDF files are then periodically updated with new evaluations based on the current state of knowledge. Cross section evaluations for a particular nuclide are performed by gathering all pertinent credible measured data, combining and comparing this with results from theoretical nuclear model computations, performing appropriate normalizations, constructing model-based interpolations and extrapolations where there are gaps in the data, and in general developing a complete data set over the incident neutron energy that is then accepted by CSWEG as the current “best estimate” standard.

In many cases the neutron cross section evaluations for nuclides of interest in reactor design and operation have not changed for a period of time and are considered to be very accurate representations of the true physical situation. Generally this occurs where dozens, if not hundreds, of independently measured data sets exist, with independently validated coverage of all neutron energy ranges of practical interest. In this case, the evaluations can properly reflect appropriate combinations of available experimental information, with a minimum of computational interpolations, extrapolations, normalizations, and other artifices.

Figure 2 shows the ENDF evaluation for the U-235 fission cross section, which is generally considered to be very well characterized. Several of the many experimental data sets available for this nuclide have been superimposed on the evaluated curve. The length of the data points in the vertical direction indicates the uncertainties in extracted cross section values. One can see that the evaluation and the measured data are self-consistent, with no gaps over the energy range covered. It may be noted for clarification that the large, closely spaced oscillations in the epithermal energy range (1 eV – 10 keV) are due to resonance phenomena. Although it is difficult to see, the cross section data do in fact closely track the evaluation in this energy range as well, albeit with some discrepancies in the exact location of the center of some of the resonances, and with some uncertainty in the top part of the resonance range.

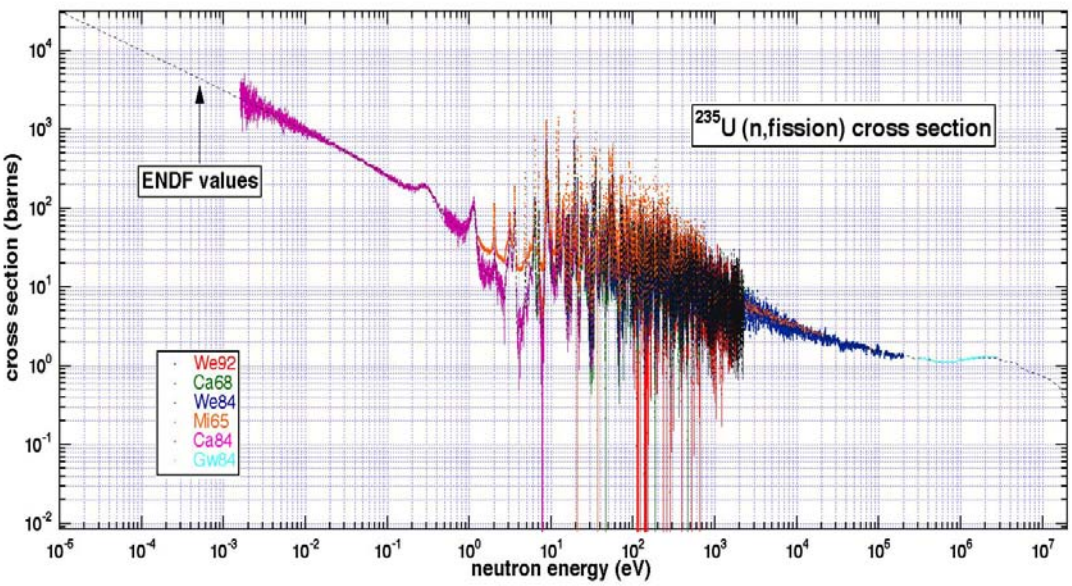

Figure 2. U-235 fission cross section as a function of incident neutron energy.

In contrast, the existing cross section data for the heavy actinides (Np-235 - Cm-250) are inadequate for the purposes of the fast nuclear reactor designs detailed in the Gen-IV Technology Roadmap and for the various purposes outlined in the AFC Program Plan. The newest generation of reactors will in many cases feature fast neutron spectra and will be designed to burn heavier actinides of neptunium, americium, and curium that are of little importance in the thermal neutron spectra of current US reactors. There is therefore a strong need for evaluated cross section data for those isotopes. When compared to the amount of cross section data available for uranium and plutonium, the quantity of data for neptunium, americium, and curium is inadequate. For instance, nearly 500 independent cross section data sets exist for U-235. These data sets comprise hundreds of thousands of individual data points. By comparison, only 30 data sets exist for Cm-244, and these contain around 7000 cumulative data points. Even fewer data sets exist for all of the other Cm isotopes of interest (Cm-242 – Cm-250).

The quality of the cross section data for the heavier actinides is also inadequate, relative to current needs. The majority of the data was acquired in the early and middle 1970's, largely during underground nuclear test explosions where the experimental conditions were essentially impossible to control. These data sets were obtained by digitizing an oscilloscope trace of the detected emissions, and typically report around 30% error although in comparison with other measurements they appear about a factor of 100 higher. The remainder of the data sets was obtained with ion and fission chambers at ORELA [Oak Ridge National Laboratory (ORNL)], lead slowing spectrometers at RPI (Rensselaer), or polymer film at Kiev. None of these detection methods achieve the standards of modern spectroscopy. For comparison, a large amount of the U-235 cross section data was measured in the 1980's and 1990's using pulsed linear accelerators, surface barrier, and germanium detectors.

It is thus quite apparent from examining the available data that the minor actinide cross sections are not known to nearly the same accuracy as the uranium and most plutonium cross sections. In fact, some of the plutonium cross sections in the energy ranges of interest are also questionable. In order to design reactors and accelerator based transmutation systems to the same (or even nearly the same) precision as current systems, these cross sections must be known to a much greater degree of accuracy.

A few examples will further illustrate the situation. Figures 3 and 4 show the fission and capture cross sections for Np-237 in the same format as was previously used for U-235. This is one of the most important minor actinides of interest for Gen-IV and AFC. It is also becoming an increasingly serious proliferation concern. There are gaps in the data, and even the most accurate measurements (the ones used for the illustrations) are not always consistent with the evaluation. As a practical outcome of this, computations of the critical mass of Np-237 performed using the currently available data produce results that vary ~±20 kg around the most recent estimated value of about 60 kg, that was derived from a replacement critical experiment performed at Los Alamos National Laboratory (LANL). In contrast, it is routinely possible to calculate, *a-priori*, the critical mass of U-235 to at least three orders of magnitude better relative accuracy.

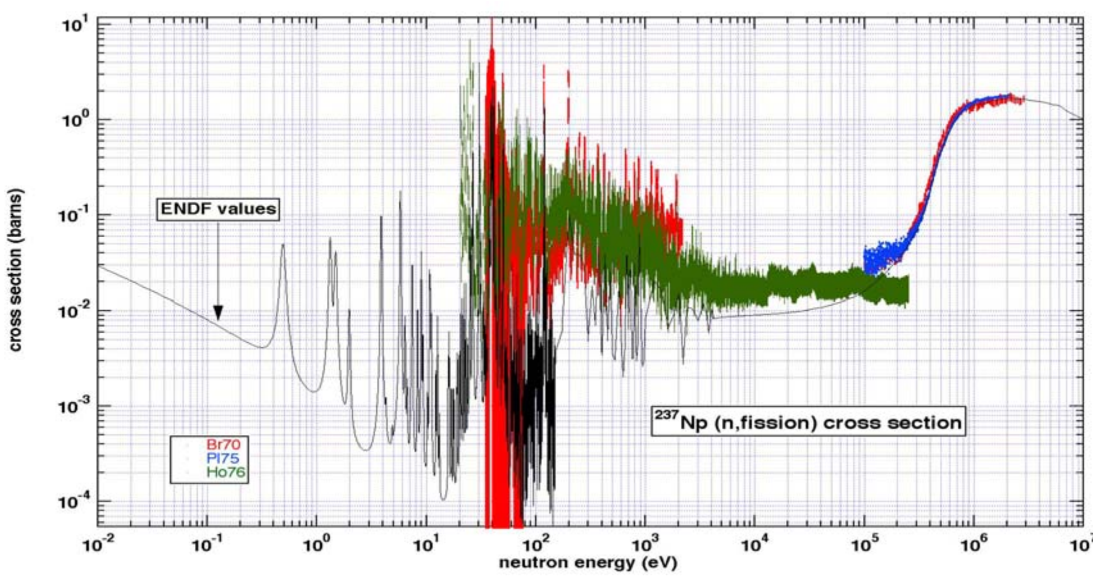

Figure 3. Fission cross section for Np-237.

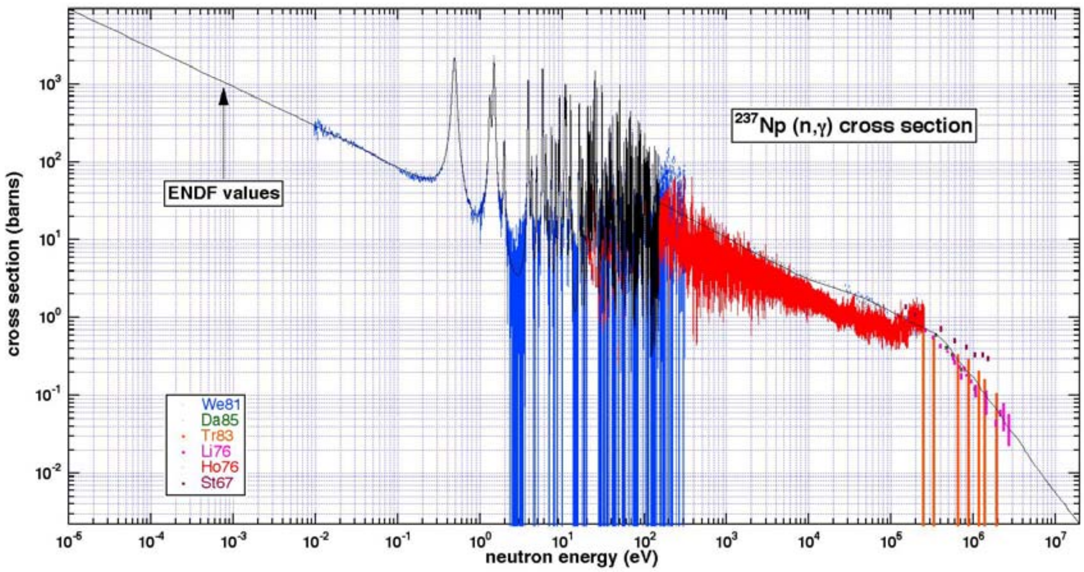

Figure 4. Capture cross section for Np-237

The fission cross section for Pu-241 is shown in Figure 5. In this case the individual measured data points for the data sets shown have small experimental uncertainties, (uncertainty comparable to the size of the data points shown, with the points simply connected by the green line) but the data points do not appear to track the ENDF evaluation particularly well. Furthermore, different evaluations (there are parallel counterparts to ENDF in other parts of the world) for the cross section of this nuclide are not in good agreement with each other, and this raises questions concerning the accuracy of engineering computations based on the different evaluations, as will be illustrated later.

Finally, Figures 6 through 9 show some cross section information for Cm-242, Cm-243, and Cm-244. These nuclides are also important in advanced systems, not only because of their reactivity effects, but also because Cm-244 becomes the primary source of decay heat at a certain point in some advanced high-burnup fuel cycles. In these cases there are several problems. The data are quite limited, the reported experimental uncertainties (again shown here by the vertical size of the various data points) are high and the currently available evaluations sometimes do not always track the experimental data. Additional plots for isotopes of curium above mass number 244 and for the important isotopes of americium are given in Appendix A for completeness. The patterns are largely the same. Clearly, there is need for improvement.

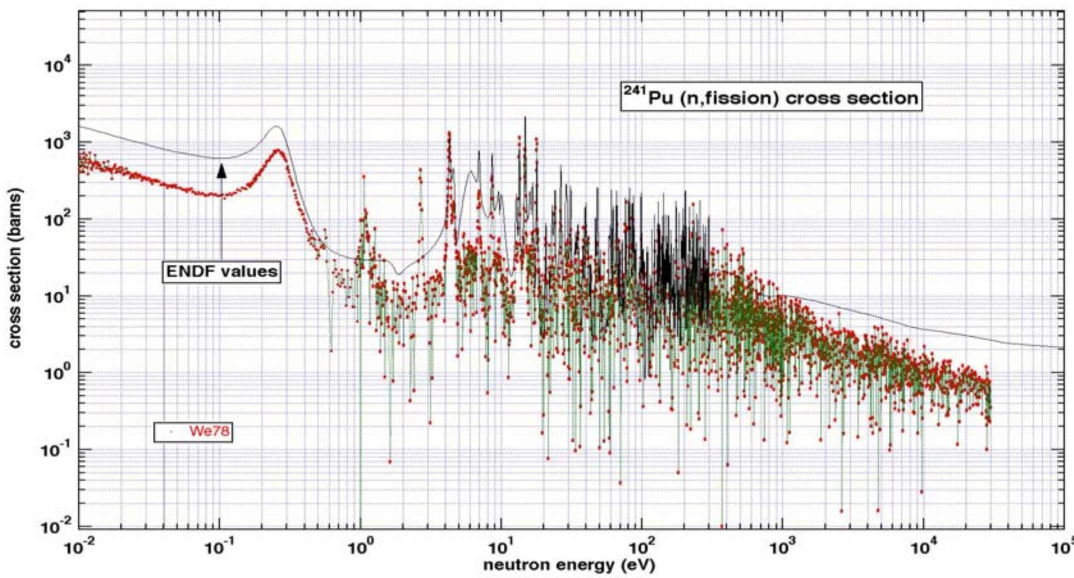

Figure 5. Pu-241 fission cross section.

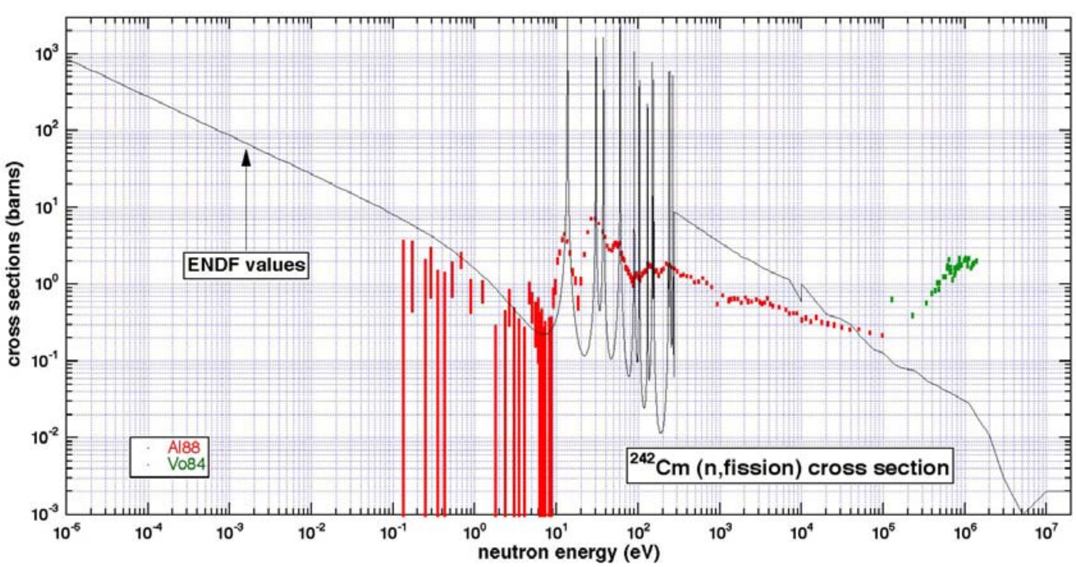

Figure 6. Cm-242 fission cross section.

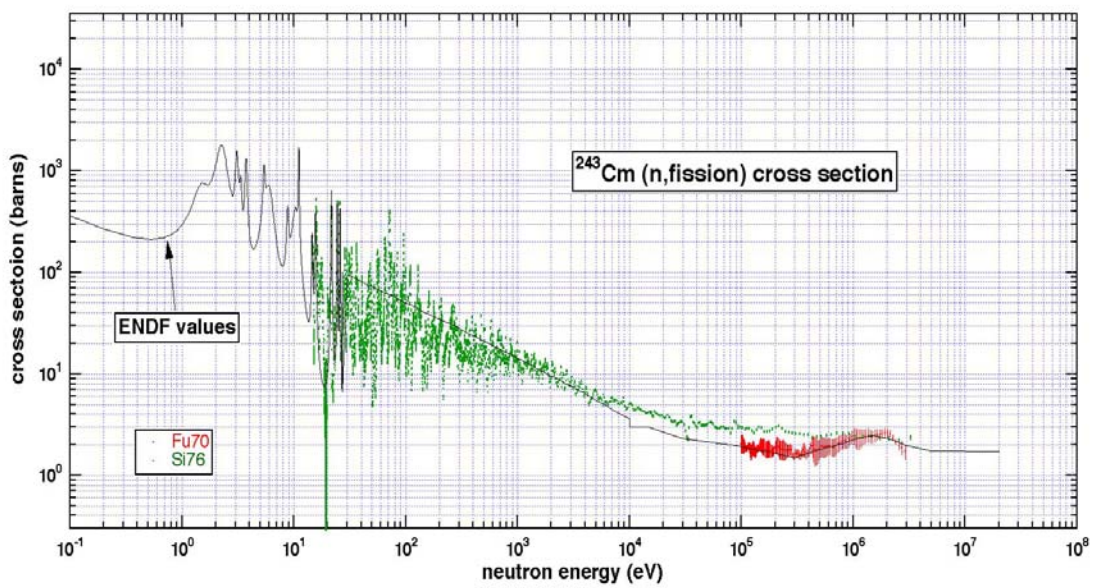

Figure 7. Cm-243 fission cross section.

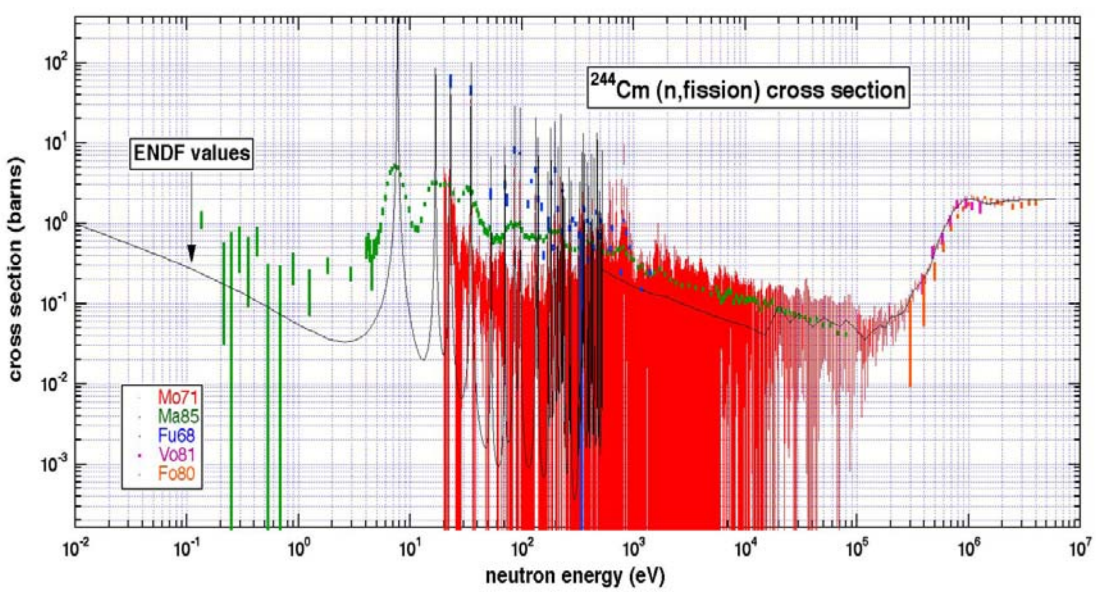

Figure 8. Cm-244 fission cross section.

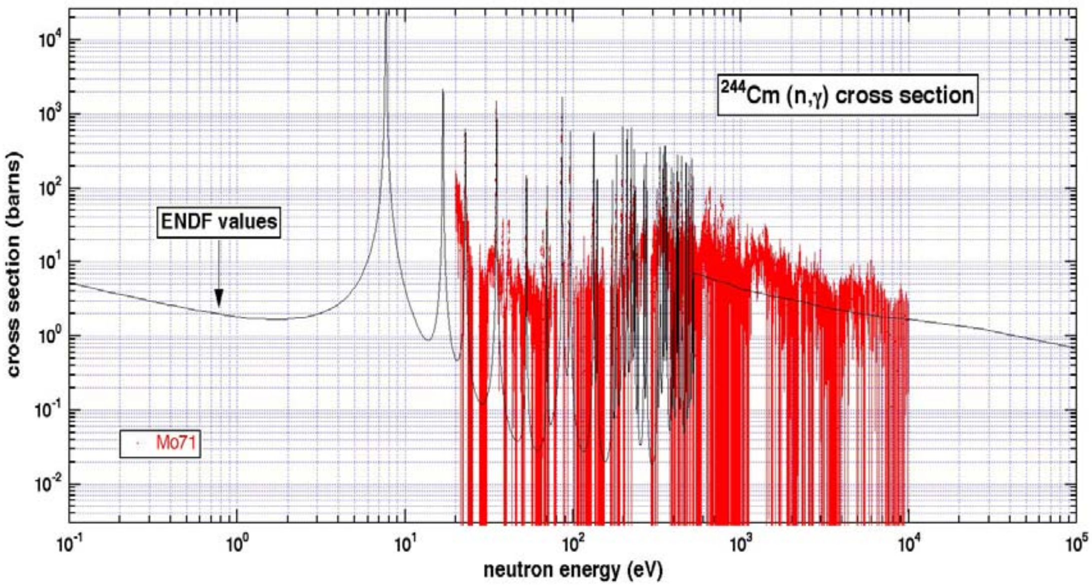

Figure 9. Cm-244 capture cross section.

2.2 Propagation of Nuclear Data Uncertainty

In this section we provide a few salient examples showing how uncertainties in the basic nuclear data can have very significant practical engineering consequence. It should be recognized that these are not contrived "worst case" situations – they are typical examples of the data issues that must be addressed in the design, operation, and maintenance of the types of reactor and accelerator systems and complex fuel cycle scenarios contemplated in the Gen-IV and AFC programs.

2.2.1 High-Burnup Light Water Reactor (LWR) Fuel Cycles

Before proceeding to the more sophisticated systems and fuel cycles associated with Gen-IV and AFC, we should note that uncertainties in transuranic cross section data are of safety and regulatory concern even with the more aggressive, high burnup, once-through fuel cycles under consideration with the present generation of LWR power plants. For example, where it is allowed, "burnup credit" in connection with criticality safety assurance of spent fuel is permitted by the US Nuclear Regulatory Commission (NRC) only for the first 40,000 MWD/MTU because of the lack of reliable data for higher burnup regimes where the minor actinides begin to play a significant role in the overall safety margin of reactivity for the fuel in question (Gauld and Parks, 2001). Furthermore, studies show that Cm 244 becomes the dominant contributor to the total decay heat in high-burnup fuel under a number of scenarios. As shown in Figure 1, Cm- 244 builds into the fuel through a complex series of interactions, each of which depends on the cross sections of the various precursor nuclides. The final expected concentration (and therefore the expected decay heat that must be accommodated) is highly sensitive to uncertainties in the nuclear cross section data for several of these precursors with sensitivity coefficients approaching

1.0 (a very high value) for some precursors (Gauld and Parks, 2001). Improved nuclear data pertinent to several of the actinide constituents of high-burnup fuel would permit correspondingly improved and cost-effective (less conservative) management of spent fuel in the higher burnup regimes.

2.2.2 Resonance Interference

Unless a reactor uses fuel that is composed of only one fissile nuclide (e.g. pure U-235 and nothing else), and no other materials having a significant degree of neutron resonance absorption behavior in their cross sections, the phenomenon of resonance interference must be considered in system design and operation. In simple terms, resonance interference causes the fission and absorption rates for each constituent of reactor fuel to be notably different (typically smaller) in the mixture than would be the case for each constituent alone, all other conditions being equal. This can have profound, often nonlinear, effects on the operating characteristics of the system. A proper understanding of these effects requires accurate cross section data and Doppler broadening parameters for the various fuel constituents. In the case of current LWR systems, resonance interference is of relatively small (but not totally negligible) significance because most of the system reactivity is due to neutron interactions in the thermal energy range. This is definitely not the case with the fast spectrum systems associated with Gen-IV and AFC, and the situation is further complicated when the fuel consists of a complicated mix of several fissile, fissionable, and fertile nuclides.

A relatively simple, but illustrative, example of how resonance interference can dramatically affect the operating parameters of a reactor is given by the Power Burst Facility (PBF), at the INEEL. The PBF was an intermediate-spectrum test reactor that operated during the 1970's and early 1980's. It was used extensively for testing of LWR fuels in a large central experiment space under extreme, sometimes destructive, conditions of power, temperature, and coolant flow. Experience with the PBF showed that, with the particular fuel that was used (18% enriched uranium), the capture resonances of the U-238 interfered with the fission resonances of the U-235 in a manner that was not as well understood as it should have been when the system was first operated in the natural transient mode. The curve of reactivity insertion vs. total energy release that was measured in the initial startup tests deviated significantly from what had been expected on the basis of the original design calculations. The negative fuel temperature feedback was greater than expected, requiring a larger reactivity insertion to produce a given desired energy release. Various investigations were undertaken to identify the source of this anomaly, which was viewed as a non-conservative situation in terms of potential safety issues. After various factors such as kinetic distortion of the flux, uncertainties in the exact control system parameters, and so forth, were accounted for as carefully as the methods of the time permitted, the problem persisted. Ultimately, an improved analysis of temperature dependent resonance interference in the fuel was required to adequately explain what was going on, and to allow for approval of the necessary revised operating procedures and safety analysis.

The PBF fuel featured a simple mixture of uranium isotopes for which very well characterized cross section information was available, and there was no significant burnup. The situation is far more complicated with the multi-nuclide fuel designs associated with Gen-IV and AFC. There will be more nuclides with very pronounced resonance capture and fission characteristics interfering with each other and the operating temperatures will extend over a wider range than is the case with current LWR systems. Furthermore, most Gen-IV concepts will have intermediate or fast neutron spectra, so more of the total reactivity will be attributable to neutron interactions in the resonance range. It will be essentially impossible to adequately understand the operating characteristics of these new systems without much-improved cross section information for the variety of transuranic nuclides that will be present in the fuel. In fact, a case could be made for the need to improve the quality of some of the current cross section data for the most well-characterized nuclides in the context of Gen-IV (e.g. U-235, U-238 and Pu-239), not to mention the more exotic nuclides about which relatively little is currently known.

2.2.3 Reactivity Management in Actinide-Burning Advanced Reactors

Several concepts for advanced fast-spectrum reactors designed to generate power or process heat while simultaneously burning actinides produced as waste in thermal spectrum reactors are currently under study in connection with the DOE Gen-IV program, as described in the recently-released Gen-IV Technology Roadmap. One such system involves bismuth and lead as the coolant, with the fuel composed largely of recycled heavy actinides. A recent computational study for this concept (MacDonald and Buongiorno, 2002) showed that uncertainties and consequent evaluation differences in the underlying nuclear cross section data can cause differences the computed system reactivity of as much as a factor of 3 or more at various points in the cycle. Figure 10, from the cited reference, shows the computed k-effective for a conceptual system design based on this reactor concept at various points in time during the burnup cycle. Two different sets of cross sections for the MCNP (Breismeister, 1992) computational reactor model were prepared for this study, one based on the ENDF/B-VI files and the other based on the European JEF files. Because all other modeling assumptions (geometry, temperature, etc.) were the same for both cases, the effects of the different cross section evaluations are isolated by this approach. As can be seen, the computed results for the two cases are dramatically different at most points in the cycle.

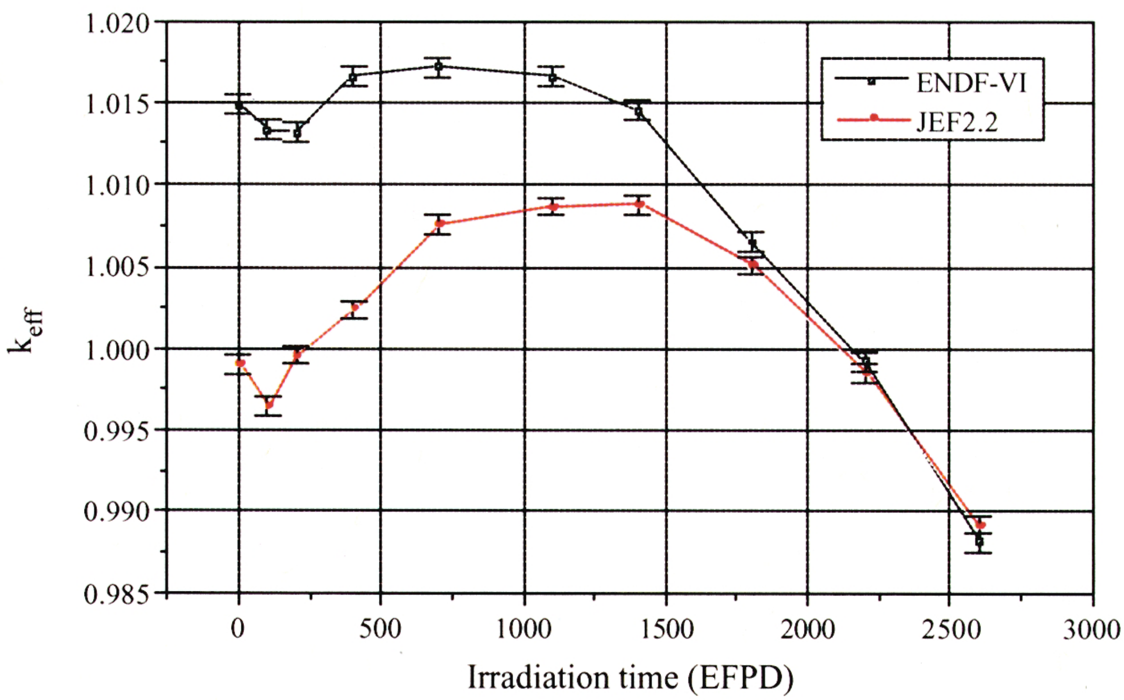

Figure 10. The effect of cross section library differences on the burnup-reactivity curve for a bismuth and lead cooled fast reactor concept.

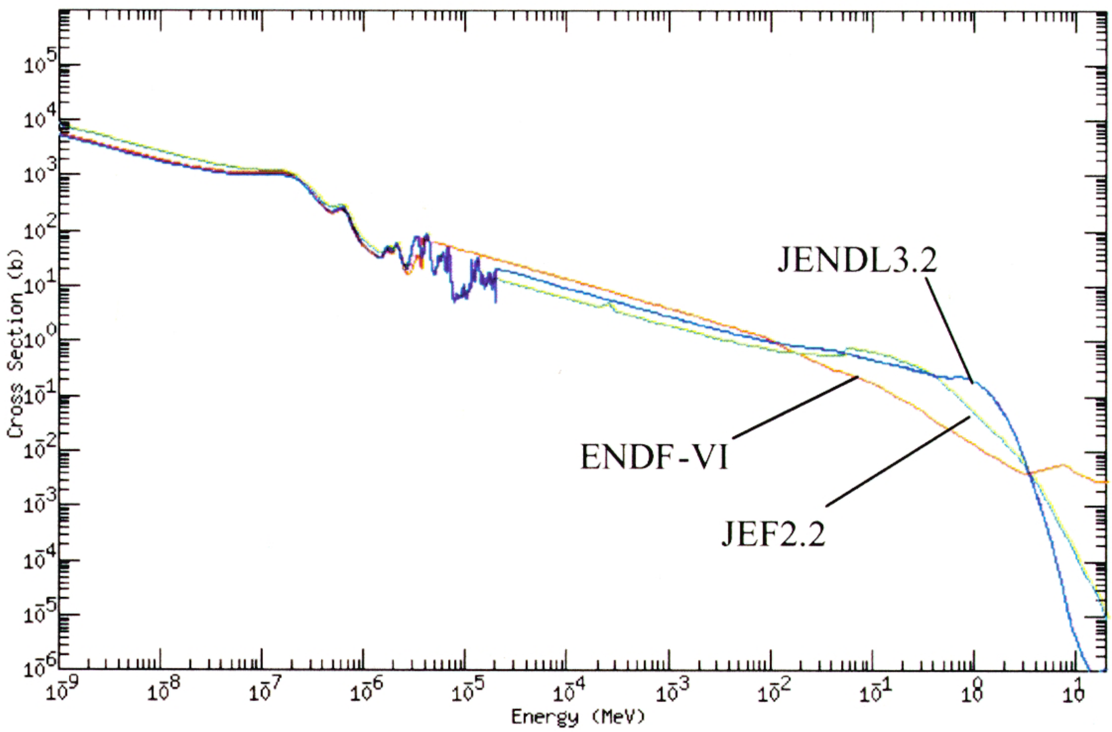

Figure 11. Am-242m Capture cross section.

An investigation into the causes of this difference, documented extensively in the cited reference, revealed that the evaluated cross sections for several important actinides were significantly different in the JEF data relative to ENDF. For example, Figure 11 shows three different evaluations for the Am-242m neutron capture cross section. They differ significantly, especially in the energy range above roughly 10 keV, which is the energy range of primary interest for most fast spectrum reactor concepts. This is not unusual for many of the higher actinides of importance for the various Gen-IV concepts. Although the low-energy evaluations are generally in reasonably good agreement among the various cross section libraries, the evaluations for higher energies vary significantly, primarily due to the limited quantity of data available, and the somewhat high uncertainty of what is available. Different evaluators will often produce different results when the quality and quantity of the underlying data are so questionable. In the case of Am-242m, capture cross section data are in fact practically non existent.

The situation described above for Am-242m is also consequential even for some of the more common nuclides such as Pu-241, as described previously and as further illustrated in Figure 12. This figure shows two different evaluations of the capture cross section for this nuclide. Once again, the evaluations differ significantly in the higher energy range.

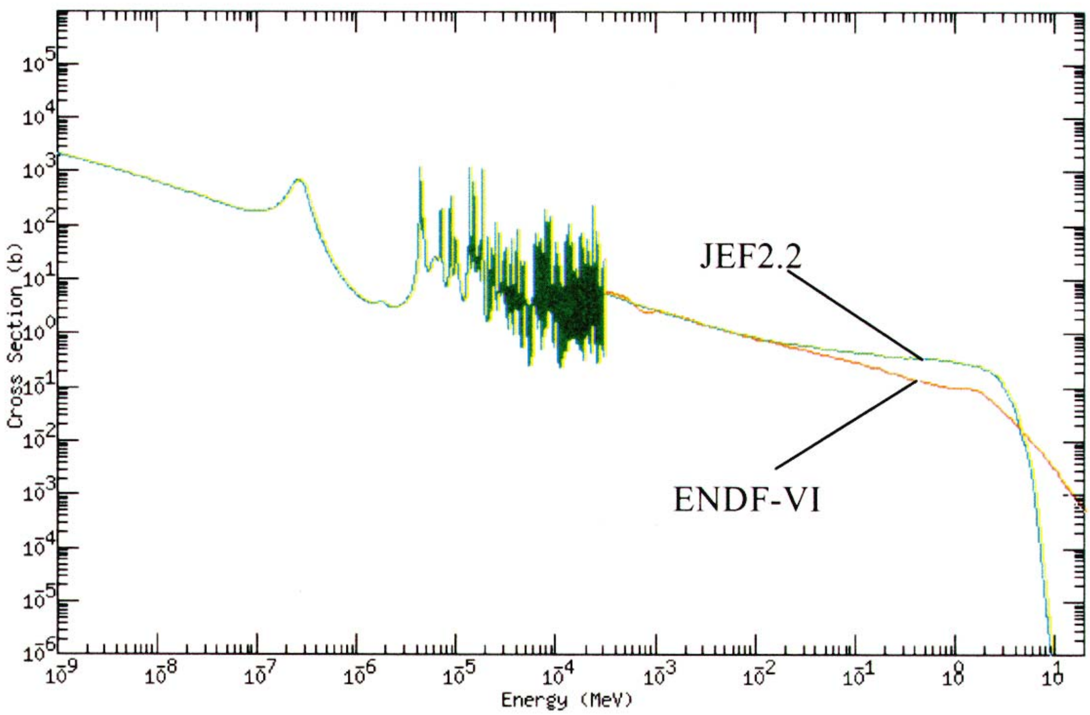

Figure 12. Pu-241 capture cross section.

2.2.4 Performance of Accelerator Driven Transmuter Systems

Development of accelerator driven subcritical assemblies for actinide transmutation is of interest in the AFC Program. In the simplest terms, these systems feature an accelerator neutron source that is coupled to a subcritical assembly in a manner whereby the neutron source is multiplied many times in the fuel-actinide assembly to produce the desired neutron flux level and neutron interaction rates in the various constituents.

The magnitude of the amplified neutron flux in an accelerator-driven system can be roughly approximated by the formula $\Phi=Ck/(1-k)$. In this formula Φ is the volume average neutron scalar flux integrated over all neutron energies, k is the fundamental mode multiplication factor for the source-free assembly and C is a constant that accounts for the source intensity, for flux shape distortions caused by the fact that the system is driven by a source and is not actually in its true source free fundamental critical mode, and other factors.

In this simple illustration it is possible to see how increased accuracy of design data (cross sections) can translate directly to significantly increased operating efficiency and corresponding reduced cost. For efficient multiplication of the accelerator source neutrons, which are expensive to produce, one typically would operate a transmutation system of this type with a subcritical multiplication factor (k) as high as possible, within appropriate safety limits. Assuming that the multiplication factor obtained from the design computations was known to within, for example, 0.5%, one might choose to operate the system with a maximum allowable multiplication factor during the cycle of perhaps 0.98. However, if the multiplication factor could only be computed to an accuracy of roughly 2-3%, as illustrated in Figure 10, then it might reasonably be necessary to operate with a maximum multiplication factor of only 0.93 to maintain the same safety margin. The resulting neutron flux available for transmutation (with the same source intensity) would therefore be reduced by a factor of almost 4, with a commensurate decrease in transmutation efficiency per source neutron, all other conditions held constant.

2.3 Current Efforts to Upgrade the US Nuclear Data Base

There has been a declining level of support for low-energy nuclear physics in the United States for the past 30 years. Although physics research in general has been sustained, both university and governmental facilities for low-energy research have been cut or completely eliminated. Cross section measurement activities in the US over the last 20 years have therefore been limited, and generally focused on the most urgent demonstrable needs in connection with nuclear criticality safety assurance, not reactor physics. There have also been some recent measurements of interest to the defense community but this is focused on uranium and plutonium isotopes, not the heavier actinides.

Although the underlying science for the various applications where cross section data are required is the same, the priorities for various types of data tend to be different, depending on the end use. In the case of criticality safety, the highest priority is currently placed on improved data in various energy ranges (largely above 1 keV) for non-fuel materials including Gd, Be, Fe, Ni, Cr, Mn, Cl, Cu, Si, Ce, Ca, Al and ZrH, as noted at the most recent DOE Nuclear Data Advisory Group (NDAG) meeting. NDAG operates under DOE-NNSA auspices to provide technical guidance on nuclear data needs to support the DOE Nuclear Criticality Safety Program, which is largely focused on stewardship of the large quantities of defense-related fissile and fissionable materials located throughout the DOE laboratory complex. There are also some stated needs for improvements in U-238 and U-233 data. The results of these measurements for non-fuel materials are of course also likely to be of use to the Gen-IV and AFC programs, although the need is not as crucial as it is for the heavy actinides.

The ORELA accelerator facility at ORNL has historically been involved in measurement of cross section data for criticality safety. ORELA was originally constructed to provide improved cross section data for the Clinch River Breeder Reactor Project. The most recent measurements performed at this facility have been for U-233, Si, Cl, Fl, and oxygen and total cross section measurements by transmission for U-235 (H. Derrien, *et al.*, 2000). Measurements of K-39 and K-41 are currently in progress. High-priority measurements planned for the near future include the capture cross section for Mn-55, Gd-155, and Gd-157. All of these measurements were, or are being, performed in support of the DOE Nuclear Criticality Safety Program.

The DOE AFC Program Plan (Revision C, November 20, 2002) describes a proposed effort, centered at the Lujan Neutron Scattering Center and the Weapons Neutron Research (WNR) facilities LANL, to improve the US cross section database for several actinides of current interest. The effort is anticipated to involve measurements of new data as well as the development of new evaluations based on existing and planned new data. The neutron energy range for the proposed measurements is stated to be from thermal to 100 keV and possibly higher. The order of priority for the various actinides as stated by AFC is given as: Np-237, Am-241, Am-242m, Cm-243, Pu-238, Pu-240, Pu-241, Cm-242, Pu-242, Np-238 and, finally, Am-242. Milligram- sized samples will be used for the measurements. These targets ar to be prepared in collaboration with the University of Nevada at Las Vegas. The planned DANCE detector at the Lujan center will be used for the capture cross section measurements. An unspecified new detector system is to be developed to measure fission cross sections. These measurements should provide some useful information, although the small mass of the target samples is likely to introduce some challenges due to the very low interaction rates that will need to be detected in the inevitable background radiation fields. Independent validation of such measurements, per standard scientific procedures, will be of importance prior to the development of new evaluations for general use.

The INEEL, using well developed, currently active, capabilities described in the following section, is in a position to perform significant additional cross section measurements for actinides of interest, and independent validation of the INEEL results can be performed using the results obtained by LANL and others. Furthermore, the measurements can be made in a manner that features, over the longer term, significantly increased accuracy and resolution, over a broader energy range extending to 2 MeV, thus covering the essentially the entire energy range of interest for Gen-IV and AFC. These measurements will compliment and improve upon, not duplicate, the work of others. The following section summarizes some technical details of the methods to be used.

3.0 INEEL CAPABILITIES FOR NUCLEAR DATA MEASUREMENTS

Four types of neutron interactions and possibly several gamma-ray interactions are important to nuclear energy systems evaluation and design. These are:

- neutron induced fission, (n,f)
- neutron capture followed by emission or one or more gamma rays, (n,γ).
- neutron elastic and inelastic scattering , (n,n′).
- neutron induced particle emission, (n,particle). The reactions include, n,nn, n,np, n,α, n,αn, and so on.

Of these, fission and capture contribute the most directly to the reactivity of systems in which the materials of interest are used, and these interactions are therefore of the greatest near-term interest.

Only three pulsed accelerator neutron source facilities suitable for state-of-the-art high-resolution cross section measurements currently exist within the United States National Laboratory System. The first of these (ORELA) was discussed previously. The other two are the LANL LANSCE facility complex, also mentioned previously, and the Intense Pulsed Neutron Source (IPNS) facility at Argonne National Laboratory (ANL). At ANL/IPNS, the INEEL, in collaboration with ANL, has developed and maintained a capability for measurements of neutron interactions and fission of actinides in a manner that represents the leading edge in this research field in terms of data quality, resolution, and ability to measure all interactions of interest simultaneously. These new methods involve arrays of primarily Compton-suppressed high-purity germanium (CSHPGe) detectors operating in coincidence mode. A detailed description of the method and examples of the results will not be presented here but the importance of the effort to nuclear energy systems will be explained and the unique characteristics of the work emphasized. Since 1993 the referenced fission studies have produced over 120 peer reviewed journal and conference papers on fission.

The experimental methods, data collection techniques, and data analysis process are the important new aspects of the INEEL work and these have been developed in the low-energy nuclear physics community over the past 25 years. Two INEEL physicists have been deeply involved in these advances. Although only a summary discussion will be presented here, two references are available to the interested reader: Ejiri and de Voigt's book on spectroscopy in nuclear physics (Ejiri and de Voigt, 1989) gives a history of early arrays of detectors, and C.W. Beausang's review paper (Beausang and Simpson, 1996) on large arrays of CSHPGe detectors brings the information to the current state of the art.

During the 1970's, the use of arrays of detectors to study both low- and high-energy nuclear physics phenomena began and has changed the experiments performed in these areas. The current state of the art in low-energy nuclear physics is seen in Gammasphere, the array of 110 CSHPGe detectors funded by DOE/SC and used alternately at LBNL and ANL Physics Divisions. As is seen with Gammasphere, the resources required for such experimental capabilities is limited to national laboratories and user facilities with substantial resources. These methods and needed facilities are beyond the resources of small groups of researchers and most university laboratories. INEEL physicists have used Gammasphere on a regular basis through a collaboration to study spontaneous fission of several actinide isotopes. The INEEL physics group, with DOE/EM funding, has also installed its own small array of 12 CSHPGe detectors

(Figure 13) and 8 neutron detectors, the supporting electronics, and a data acquisition system on a beam line at the ANL/IPNS facility to study neutron induced reactions on heavy actinides. This small array is unique in the world for the capability of studying neutron-induced reactions in the new way.

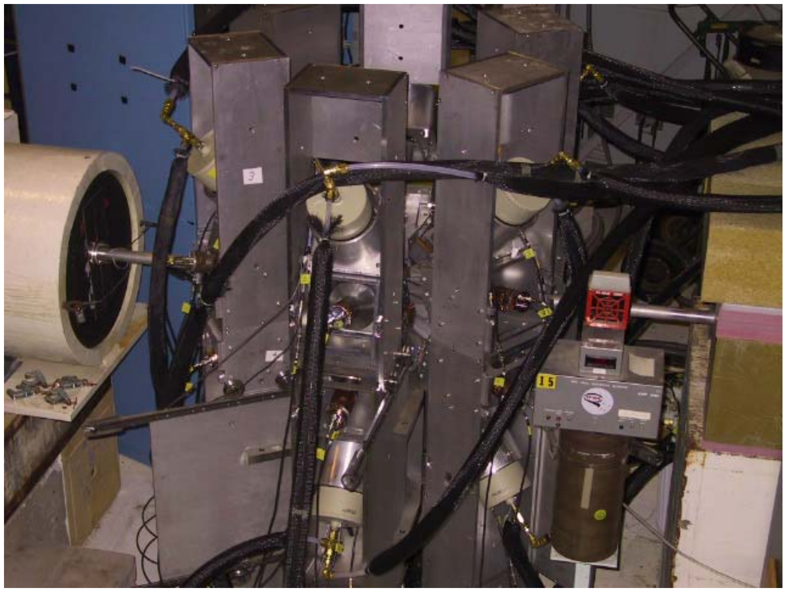

Figure 13. INEEL Detector array at ANL/IPNS.

The new methods developed for operating arrays of CSHPGe detectors in fast coincidence mode, acquiring data event-by-event (a single neutron interaction is an event), and using advanced data sorting is providing both new and better results than have ever been obtained previously. A brief overview of certain key aspects of the experimental apparatus and procedure is given below. Additional details are provided in Appendix B.

Compton suppression is the method of rejecting radiation that strikes a detector but does not deposit all of its energy in the detector. Although the method gets its name from the rejection of Compton scattered gamma rays in a germanium detector, this is a method of active shielding that works with detectors for different radiation. In the case of germanium detectors, the technique is to surround the germanium crystal with a second scintillator that has a high probability of detecting the gamma ray that scatters out of the germanium crystal. A good Compton suppression system will reduce the Compton background in a spectrum from the germanium detector by ~60%. This is a significant reduction that leads to a reduction in the statistical error of a gamma ray observed above this background.

Grouping the CSHPGe detectors into an array that is operated in coincidence mode has the additional benefit of selecting events in the experimental process that have multiple and simultaneous emissions of radiation. The early detector arrays used this method as a multiplicity filter exclusive of other selection methods. The culmination of this process was seen in the Heidelberg-Darmstadt crystal ball and the ORNL spin spectrometer (Ejiri and de Voigt, 1989) and was given a mathmatical basis by a Dutch student (Ockles, 1978) and a Danish professor (Werf, 1978) in independent works. These coincidence arrays actively reject background radiation and other time-uncorrelated emissions of radiation from the data, further decreasing backgrounds and reducing statistical error. For modern arrays this has been taken a step farther by using CSHPGe detectors with excellent energy resolution.

List-mode data collection and subsequent data sorting and extraction of the desired cross sections, discussed in Appendix B, use the high resolution of the germanium detectors to further reduce statistical error and to select subsets of data from the experimental data stream that are related to one process, i.e., n,f or n,γ processes for example. The desired cross section information can then be extracted from these data subsets.

This experimental method results in self-consistent data sets for all types of neutron interactions occurring. This means that the number of measurements needed to produce the required information is greatly reduced and cross sections for all interactions that produce measurable data can be extracted from the data sets. With the data collection method being used, the post analysis separates the data such that the data sets are as if separate experiments have been done with cross contamination at a minimum. This has the advantage of reduced experimental time as compared to running multiple experiments, consistent experimental configuration to reduce systematic errors and reduction of statistical error by rejecting and removing backgrounds. The challenges associated with these new methods include: 1) large and complex setups of experimental systems, 2) sufficient experimental run time with neutron beams to have adequate statistics and, 3) the existence of only a few neutron source facilities that will provide both intense beams of neutrons and long-term location of the experimental setups.

As with any set of experiments, the experimental methods and equipment should be consistent with the capabilities of the experimental facility, in this case the accelerator neutron source. For a multi-parameter, array based system such as the one being used at ANL/IPNS, a neutron flux of 10^5 to 10^6 neutrons per second per square centimeter induces a reaction rate that yields a few thousand (from ~1000 to ~5000) observed events per second. This rate is sufficient for study of actinides with high to moderate cross sections and low specific activity. For cases where the cross sections are low or the sample activity is high, a stonger neutron source than the current beamline at ANL/IPNS is needed, and in fact can be developed at the same facility as will be discussed later.

Finally, the INEEL has also been involved in inelastic neutron scattering experiments carried out in collaboration with Lawrence Livermore National Laboratory (LLNL) and LANL using the GEANIE detector system installed at the Los Alamos Neutron Science Center (LANSCE). GEANIE consists of 20 CSHPGe detectors and is primarily utilized to study neutron-induced reactions. At LANSCE, neutron energies from 250 keV up to 600 MeV are available. The INEEL/LLNL/LANL collaboration at LANSCE has not been as longstanding as the effort at ANL/IPNS and the resource level has been lower, but it has nevertheless produced significant useful results.

4.0 RECOMMENDED ACTIONS

The INEEL, in collaboration with key domestic and foreign partners, is well poised to make a major contribution to improve both the quantity as well as the quality of basic nuclear data required for all aspects of the Gen-IV and AFC programs. The planned effort will compliment, not duplicate, other related efforts that are ongoing elsewhere, and multiple measurements with different methods will provide the necessary checks on all data. The U.S. National Nuclear Data Program works best if there are several sets of experimental results to be compared and evaluated into a useful file. In many instances the INEEL can provide cost-effective key information that either will not, or simply cannot, be obtained by other known measurement projects. As has been stated, the availability of facilities that can produce high quality beams of neutrons over the needed energy range is limited in the United States.

There are three components to the proposed INEEL experimental effort. The first involves a redirection of the current activities at ANL/IPNS. A Np-237 target is currently at ANL/IPNS and with near-term funding to configure the beam line to allow the use of 2 MeV neutrons, some initial measurements could begin in FY-04. Work for other high-priority actinides can then follow, because the INEEL currently has a source of other heavy actinide targets of high quality and possible near term delivery. Second, additional near-term measurements for other materials of interest can also be included via the separate ongoing collaboration at LANSCE. This could be a part of the planned cross section measurement activities described in the AFC program plan, with INEEL assistance in data collection and evaluation. Finally, in the longer term, a simple steady-state neutron source facility with an array of detectors can be constructed at the INEEL to provide a very cost-effective approach for ongoing measurements of many materials of interest as issues come up in the future, and for use as a dedicated training facility to introduce the next generation of students to state of the art methods for this type of measurement.

In addition to the experimental work described above, which constitutes by far the majority of the overall program outlined in this document, there are two related supporting activities that are needed. The first of these involves a modest effort focused on the maintenance of INEEL capabilities and computational tools for processing of the basic ENDF/B evaluated files into local application-specific multigroup and few-group libraries suitable for use in physics design codes and fuel cycle analysis codes. This is an essential core capability, but it is one that has languished for some time due to lack of funding. Second, there should be an effective mechanism for communication and information interchange to ensure that the various US efforts related to NE nuclear data needs compliment each other. Although the data measurement and validation program summarized in this document would constitute one of the most comprehensive efforts of its type in the US as well as in the world, there are, as noted previously, complimentary activities of the same general nature that are either ongoing or planned elsewhere. The currently planned series of Generation-IV Nuclear Data and other topical workshops offer just such a mechanism.

The following sections provide some additional technical detail for the various experimental and computational components of the recommended INEEL effort in the area of reactor-physics-related nuclear data measurements and computational processing capabilities to support the DOE-NE mission.

4.1 New Cross Section Measurements at ANL/IPNS

The previously described experimental measurement system at ANL/IPNS is unique, with a number of attractive features. No other neutron source facility has an array of CSHPGe detectors running in coincidence mode, with the associated electronics and data acquisition system that can be dedicated full time to the proposed measurement effort. The neutron-induced fission cross sections and the prompt isotopic yields of the fission products can be determined for any fissile target that can meet radiological requirements. The neutron capture cross section (n,γ) can be determined for actinide targets with γ-rays of energy above the x-ray region. A flux of approximately 10^6 neutrons per square cm per second is currently available in a tightly focused and well-shielded beam. Very long runs are possible, up to 27 weeks of beam time per year on a 24/7 basis, and the background radiation levels are low.

In the current configuration, the neutron energy range at ANL/IPNS is ~0.001 eV to 300 eV, but this can be increased. This would first involve a change to the beamline and the detector setup to allow an expanded range of measurements, and to continue with the existing equipment until an upgrade can be obtained. By increasing the beamline length to provide an 18 to 20 meter distance between the target and the detector array, neutrons having energies up to about 2 MeV can be used. .

The recommended plan for activities at ANL/IPNS in the short term seeks to maintain and utilize the current capabilities until the described changes can be made and an even better measurement systems can be brought online. The current setup requires a few repairs, and modifications will need to be made to the IPNS experiment plan and safety documentation to accommodate new targets. Also an array of eight liquid scintillator neutron detectors at the target position needs to be incorporated back into the experimental system. A special fission chamber is being built at the Dubna laboratory that will be used as a neutron beam flux monitor. The appropriate time to incorporate this chamber is during the move of the equipment from a distance of 12.5 meters from the neutron-producing target to a distance of 18 to 20 meters from the neutron-producing target ANL/IPNS.

The addition of the neutron flux monitor and the neutron detector array will greatly enhance the capabilities of the measurement system. The flux monitor will make it possible to determine absolute cross sections and to measure the energy spectrum of the incident neutron beam. The system already has the capability to record the incident neutron energy on an event-by-event basis. The addition of the neutron detector array at the target position will make it possible to gate the coincidence data on the number of neutrons emitted from the reaction and in a very rough way on the emitted neutron energy. Once these initial system modifications are complete, calibration measurements using encapsulated high-purity U-235 targets (currently available at the INEEL) can be conducted. Some additional initial measurements at ANL/IPNS could also be conducted for Np-237. The U-235 and Np-237 measurements will provide an evaluation point for the work with the new methods described here. Simultaneously, the process for obtaining additional targets from an established source in Russia for measurements of other actinides of interest can be started. New targets would include, in approximate priority order, Pu-241, Am-242m, Np-236, Np-237 (an improved target relative to the one now on site), Pu-242, Am-243, and curium isotopes of interest.

With a near-term start, calibration data for U-235 could be available in mid to late FY-04 to judge the (n,f) and (n,γ) results and have concrete information to plan an upgrade at ANL/IPNS. This upgrade would be a gamma-ray detection system with 36 CSHPGe detectors to replace the present 12-detector array. This larger array will permit improvement of data quality, and it will significantly reduce the time required to produce a given level of statistics in the data. The 12-detector array would then be returned to Idaho for eventual use in another facility that is described in Section 4.3.

Finally, it is recommended that the upgraded 36-detector system (or a duplicate of it) ultimately be located at a more intense dedicated beam line available at IPNS, the so-called "zero degree" port. This would provide a higher neutron intensity at modest additional cost and would yield a very useful, dedicated capability to conduct high quality nuclear measurements for DOE/NE needs for several years. In the longer term, a dedicated beamline for cross section measurement in some of the most challenging cases might possibly also be established by DOE-NE in consultation with the DOE Office of Science at ORNL/SNS. This new facility will feature a neutron source intensity that is from 500 to 1200 times the intensity of any existing neutron spallation facility in the world. The only system that would rival ORNL/SNS is the CERN n-TOF facility (CERN, 2000).

4.2 INEEL/LANSCE Collaboration

As noted previously, the INEEL has maintained collaboration at LANSCE for the purpose of gathering neutron-induced gamma-ray cross section data for use in EM applications. These measurements are very similar to the types of cross section measurements needed for Gen-IV and AFC Initiative. This collaboration could easily be expanded to include additional measurements of interest and could possibly be combined with the planned LANL AFC cross section measurement campaign described previously. This would offer a relatively low-cost mechanism to produce some of the needed data in the near term. The most likely neutron source for these measurements would be the WNR facility at LANSCE. There are several measurement options available at the WNR facility. Some measurements can be done using the GEANIE array. GEANIE is a gamma-ray detector array constructed for the purpose of measuring neutron-induced cross sections in actinides, specifically plutonium. It is generally highly subscribed and it would be unlikely that a majority of the available time could be obtained. It would most likely be used for shorter, higher-profile measurements. Apart from the GEANIE array, there are other available beamline locations at WNR. A small array of detectors could be located at one of these. This arrangement could likely be maintained indefinitely for relatively little cost, and it would allow for a number of longer, or lower-profile, measurements. In addition to the WNR facility, LANSCE also contains the Lujan Scattering Center. Providing thermalized neutrons, the Lujan center is the home of the DANCE detector array mentioned earlier in relation to the LANL AFC work. This could provide another location for cross section measurements.

The primary difference between the ANL/IPNS facility and the LANSCE facility is the neutron energy. Neutron energies available at WNR extend from about 250 keV to over 500 MeV. The Lujan Center provides neutrons in the meV energy range. Of course, neutrons above 10 MeV are of limited interest in reactor or transmuter design work. While this does mean that LANSCE cannot be the only measurement location, it does mesh nicely with the neutron energies available at IPNS. Using the two facilities together, it will be possible to study neutron cross sections over the entire energy range of interest.

One benefit of collaboration with LANL and LLNL is found in the overlapping interests of weapon and reactor designers. Many of the same isotopes are of interest to both communities. Different specific interaction cross sections may be desired for a given material, but it is often the case that many different cross sections can be measured in the same "run," as described previously. This could lead to substantial cost-savings for target acquisition, equipment purchases, etc.

In summary, maintaining INEEL participation in the collaboration at WNR would be very valuable to DOE-NE. Significant contributions could be made toward reducing the backlog of needed cross-section data. Facilities exist for performing (n,γ), (n,xn), (n,f), (n,total), and (n,particle) measurements. All of these could be used to contribute to the database of design-critical cross sections.

4.3 INEEL Cf Facility

Finally, it is recommended that planning and detailed feasibility studies be initiated for the future construction of a small Cf-252 neutron source based system at INEEL that would primarily be for the measurement of (n,γ) reactions on non-actinide targets, where the backgrounds are low and there is less need for sophisticated background suppression techniques. This concept is shown in Figure 14. The NRC-licensed Cf source, with an activity of approximately 1 Curie, would provide neutrons in the fission energy range of a few keV to ~10 MeV, consistent with the spectrum in the fast-spectrum systems under consideration for Gen-IV. The measurements to be carried out in this system are singles measurements. For these measurements the only parameters that will be collected will be the time-of-flight of the neutron emitted from the Cf source and the gamma rays emitted from the interaction. The neutron time-of-flight will be determined by measuring the time difference between the gamma rays emitted by the fissioning Cf-252 nuclei and the arrival of the neutron at the target position. The Cf-252 source is located at one end of the shielded enclosure. A set of detectors is used to detect the gamma ray emission following the fission of the ^{252}Cf nuclei. These detectors are used to start the time-of-flight clock. The clock will be stopped when a gamma-ray event is detected by the Ge detectors positioned around the target sample. This will provide the time-of-flight of the neutron and therefore its energy. Initially the system could be operated with a small number of Ge detectors. The time required to collect sufficient counting statistics for a particular sample scales linearly with the number of detectors present. The current detector array from ANL/IPNS can be used once the upgraded array is available at ANL/IPNS.

The advantage of this idea is that the system would be relatively simple and inexpensive, but it will still provide useful information for some nuclides. For example, the Gen-IV Technology Development Roadmap recommends the development of improved cross section data for bismuth and lead in certain energy ranges. The recommended system could be used for this application in a very cost-effective manner. In addition, the previously cited NRC report (Gauld and Parks, 2001) noted deficiencies in the neutron absorption fractions not only for actinides, but also for various long-lived fission products, burnable neutron poisons, and structural materials, particularly Cs-133, Cs-135, Tc-99, Ru-103, Pd-155, Eu-155, Sm-150, Sm-151, Ag-109, and Mo-95, and several others also are of interest (Gauld and Ryman, 2001, Oblozinsky, 2002).

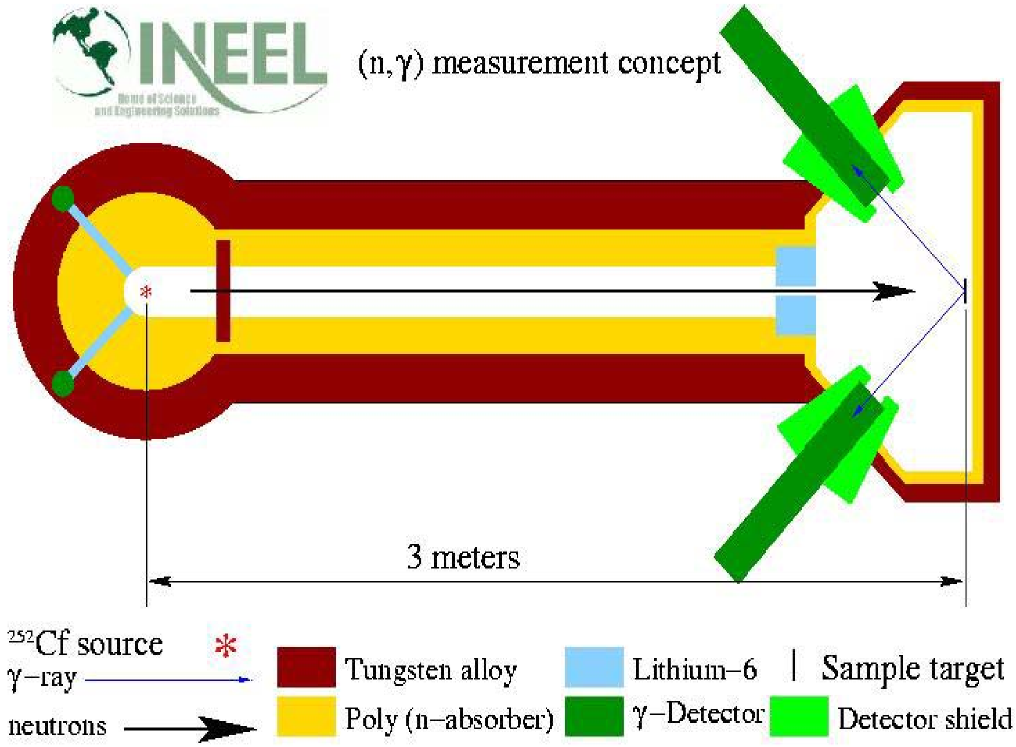

Figure 14. INEEL Cf source facility concept.

The (n,γ) resonance and background cross sections for these materials and others could be measured for a relatively low cost with the proposed system for neutron energies between a few keV and 10 MeV. The steady state nature of the proposed system and the fact that it doesn't require an accelerator based neutron source means that the individual measurement series could easily be run as long as needed to produce the desired statistical accuracy for the measurement in question. This system will not only be used for data measurements, but it also offers a very convenient low-cost facility for training of students and in-house development of electronics for upgrading the capabilities of the deployed measurement systems, without having to deal with time pressures and conflicting uses, as is sometimes the case at the various multi-user accelerator facilities.

4.4 Data Processing Code Maintenance

Finally, it is strongly recommended that there be a rejuvenated effort directed toward the improved maintenance and future development of the INEEL processing codes that are required to produce useful continuous-energy and multigroup case-specific cross section libraries for the various physics design codes. The INEEL uses a local adaptation of the well-known NJOY (MacFarlane and Muir, 1994) code for the first part of this process. The local version has been modified to include parameters required for computation of case-specific resonance self-shielding when multigroup libraries are to be produced in a second step before the information is passed to the design codes. The second step involves the use of the COMBINE (Grimesey et al, 1991) code developed at the INEEL. A version of COMBINE for processing of ENDF Version 5 data has been released via the Oak Ridge Radiation Shielding Information Center, and there have been well over 100 requests for copies. It is a very convenient tool for a variety of applications where multigroup and few-group resonance shielded cross section libraries are needed. However, the updates to COMBINE for ENDF Version 6 and the associated documentation have not been made publicly available due to lack of funding. As a service to the community by the designated nuclear lead laboratory, these software packages should be more systematically maintained and documented for periodic release. The labor cost involved would be very modest, and formal programmatic support for this activity would also allow the INEEL to participate much more actively in CSEWG than is now the case.

5.0 CONCLUSIONS

There is a substantial need for extensive new measurements to provide the neutron interaction cross sections required for long-term success of the DOE/NE AFC and Gen-IV programs. The need for improved data involves some stable isotopes and as well as isotopes of neptunium and heavier actinides. This document offers the basic outline of a plan to establish the necessary research, in collaboration with others, which will produce the necessary information.

The proposed effort thus involves: 1) a redirected continuation of current INEEL capabilities for measuring neutron interaction data at ANL/IPNS, with an intermediate-term instrumentation and beamline upgrade to provide much-improved data quality and statistical accuracy, 2) continuation of the current collaboration between INEEL and LANL at LANSCE to provide additional data in the near and intermediate term and, 3) the longer-term development of a simple Cf-based neutron source facility at the INEEL for certain types of measurements that do not require sophisticated pulsed accelerator neutron sources. The overall campaign would not only involve other DOE laboratories, but also university facility and students, as well as international collaborators. It would be the core of a renewed capability that has largely been lost in the low-energy nuclear physics community and in reactor physics work.

The building of a new accelerator neutron facility, as was done for the Clinch River Breeder Reactor program, is not part of these plans. Existing DOE capabilities will be used, with the recommended upgrade of the arrangement at ANL/IPNS and possibly incorporation of some measurements in the longer term at the new DOE ORNL/SNS facility when it comes online.

Although not specifically mentioned, this program will use university collaborators to support experimental work and data analysis. This is based on the model of the current INEEL/ANL/IPNS collaboration which has been in existence for ~10 years. The effort in this area would be to expand the collaboration and also help establish university research programs to support DOE/NE nuclear science needs in the future.

The program is to provide the highest quality results, reviewed by peers in the scientific community though publication, and an infusion of this new data into the ENDF files through the evaluation and acceptance by the U.S. NNDC.

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Appendix A

Cross Section Data References and Additional Data Plots of Interest

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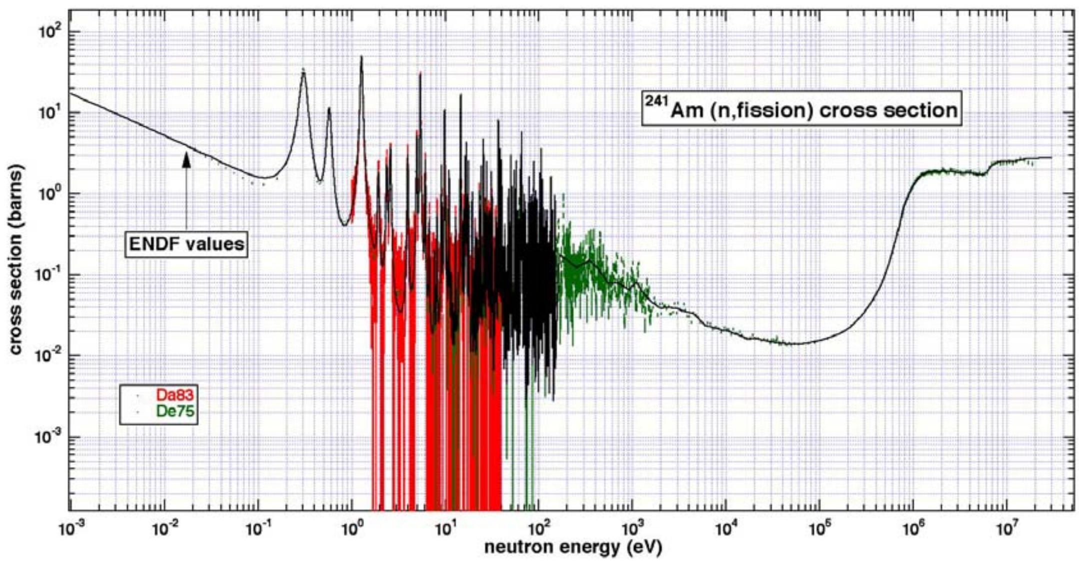
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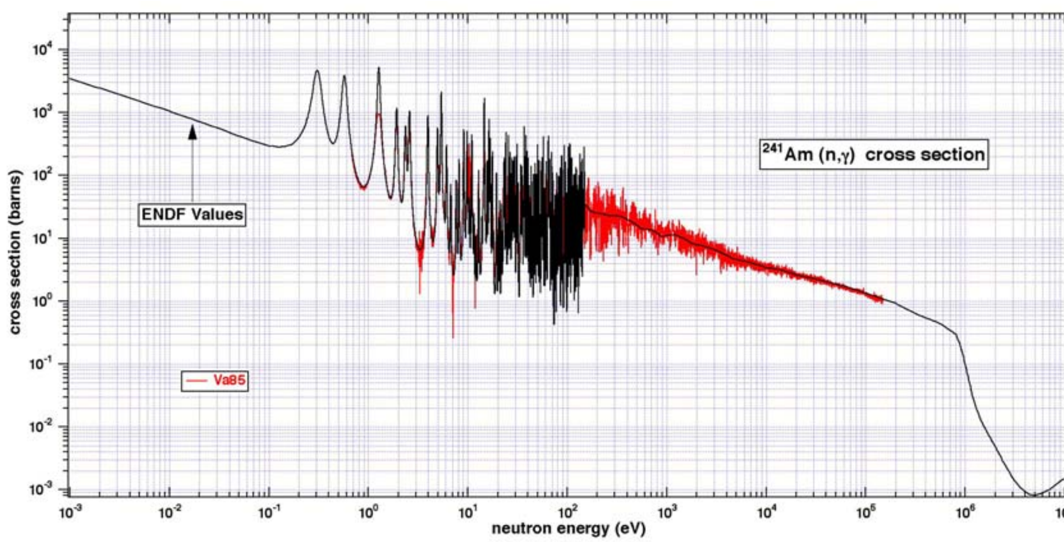
Da83
De75
cross section (barns)
neutron energy (eV)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ENDF Values
Va85
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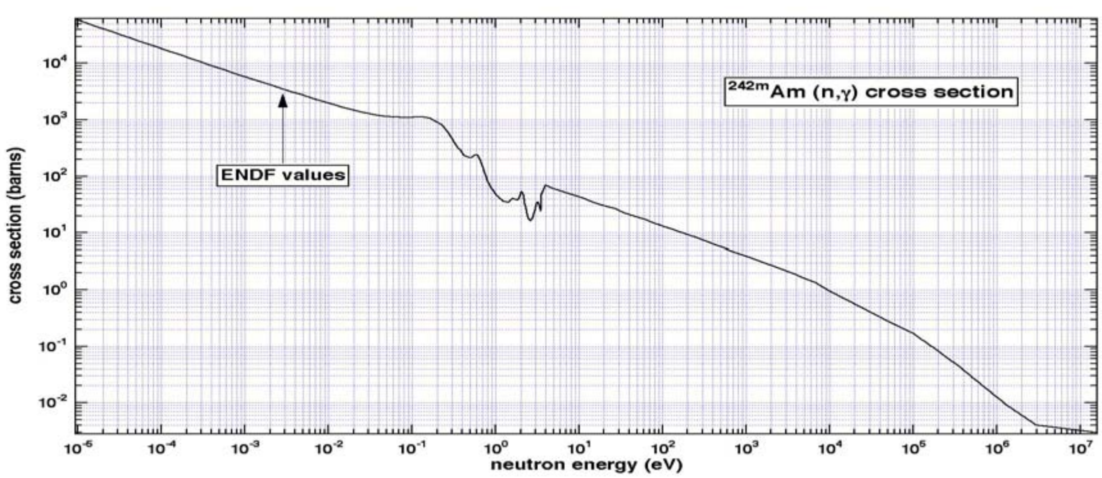
neutron energy (eV)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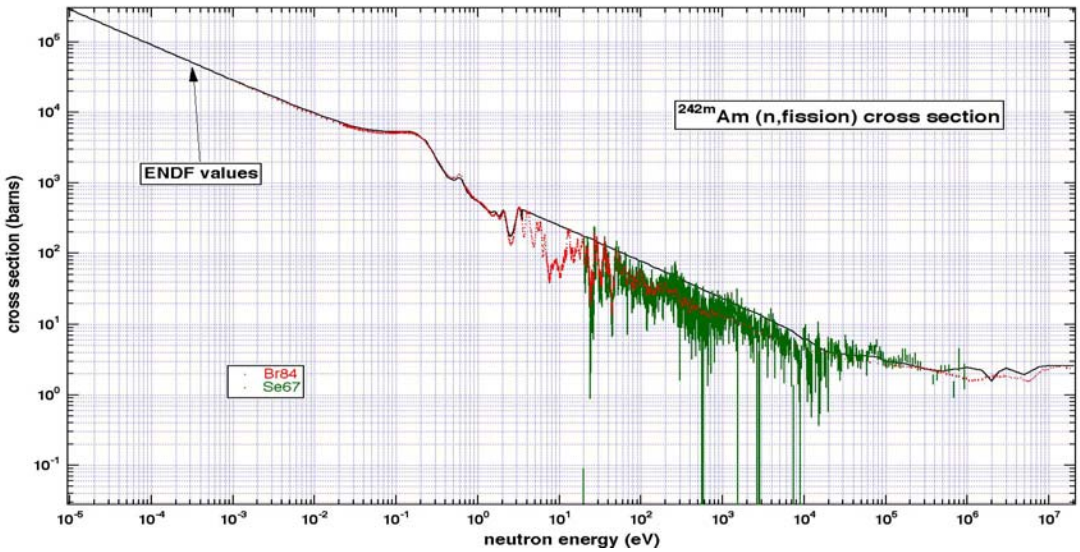
cross section (barns)
neutron energy (eV)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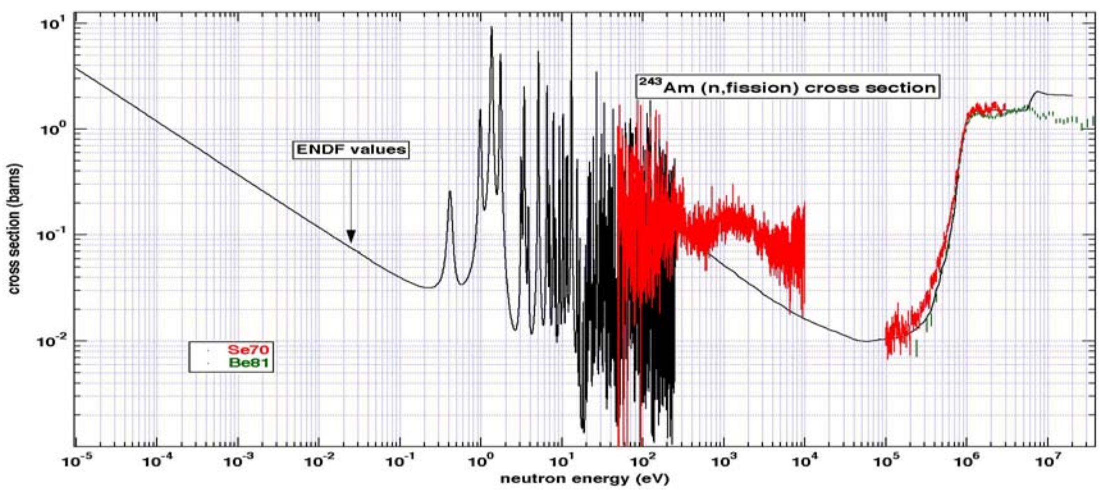
neutron energy (eV)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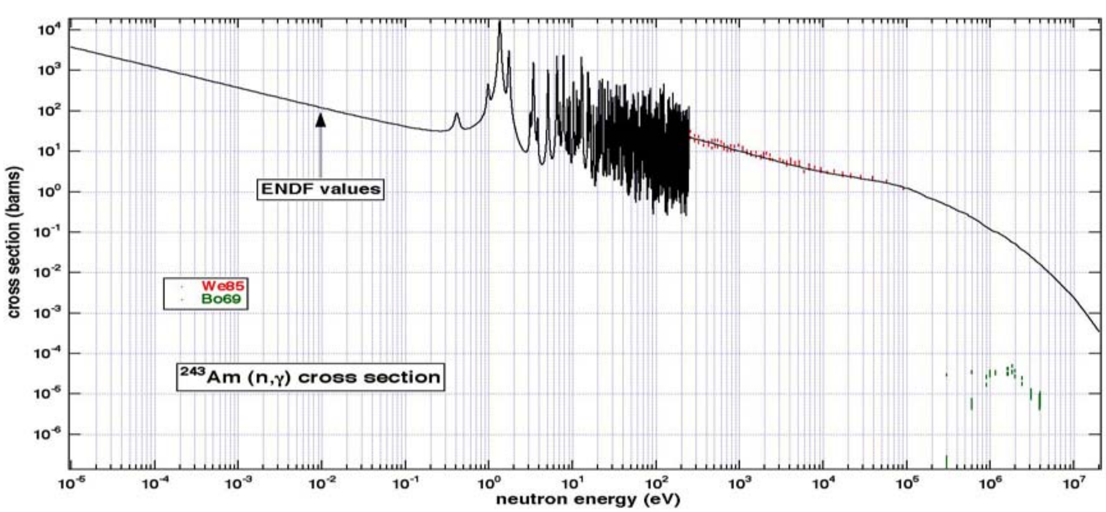
neutron energy (eV)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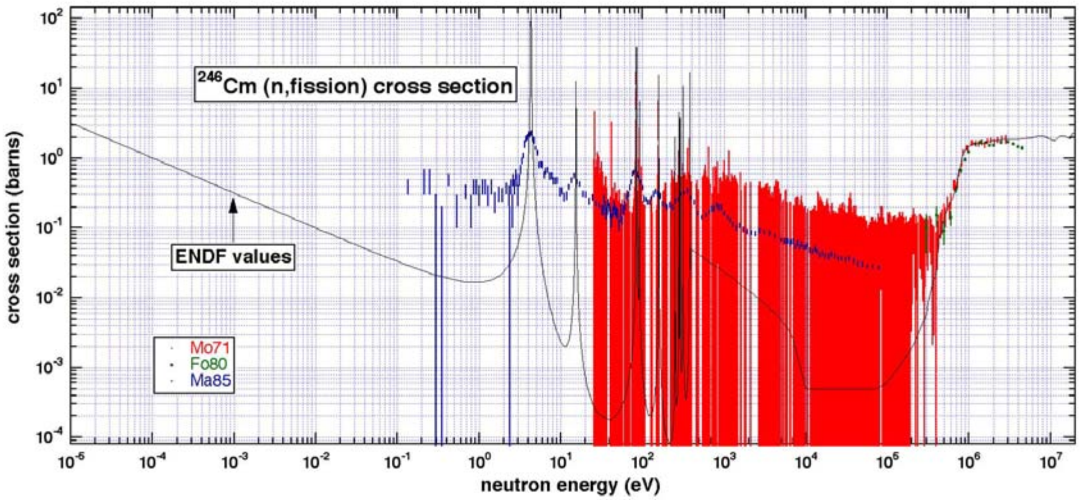

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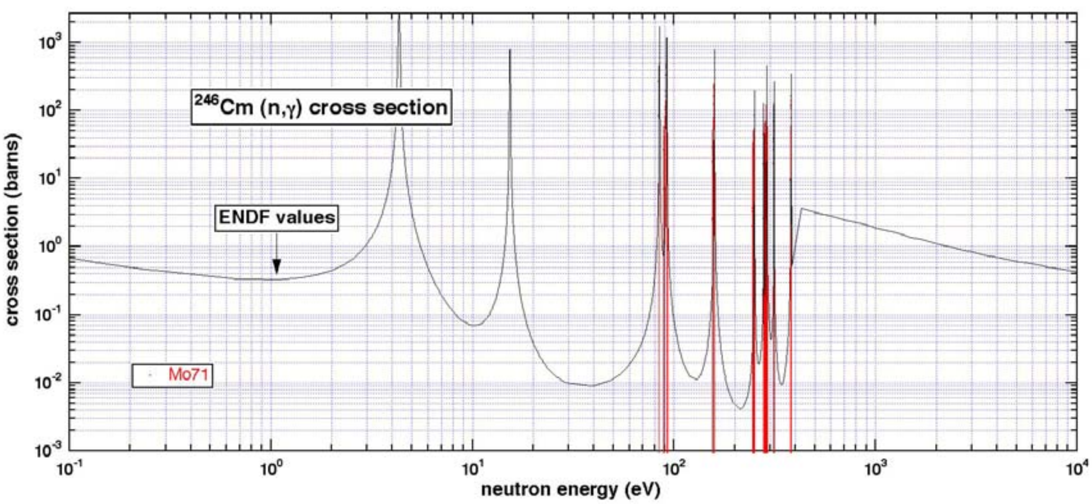
ENDF values
Mo71
Fo80
Ma85
cross section (barns)
neutron energy (eV)



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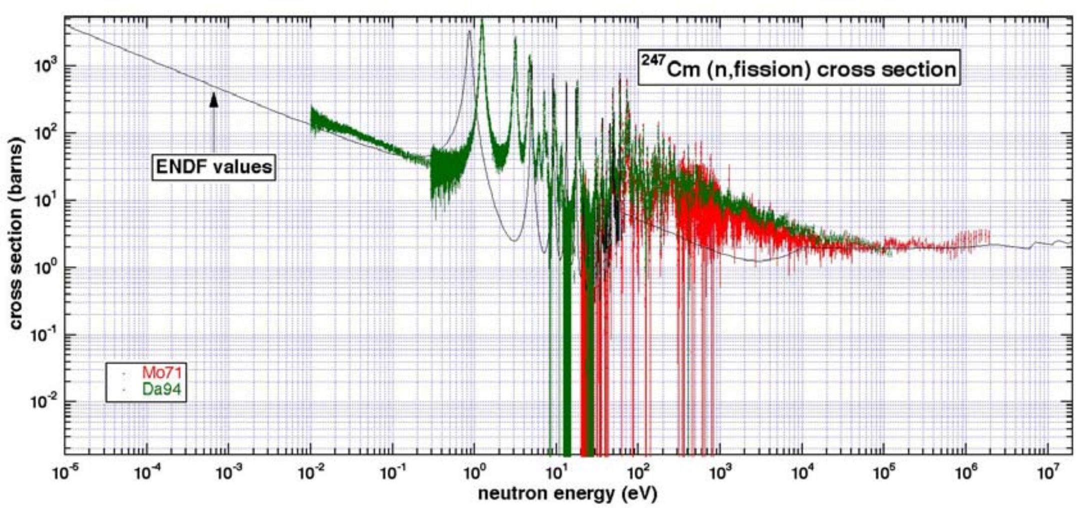
cross section (barns)
neutron energy (eV)



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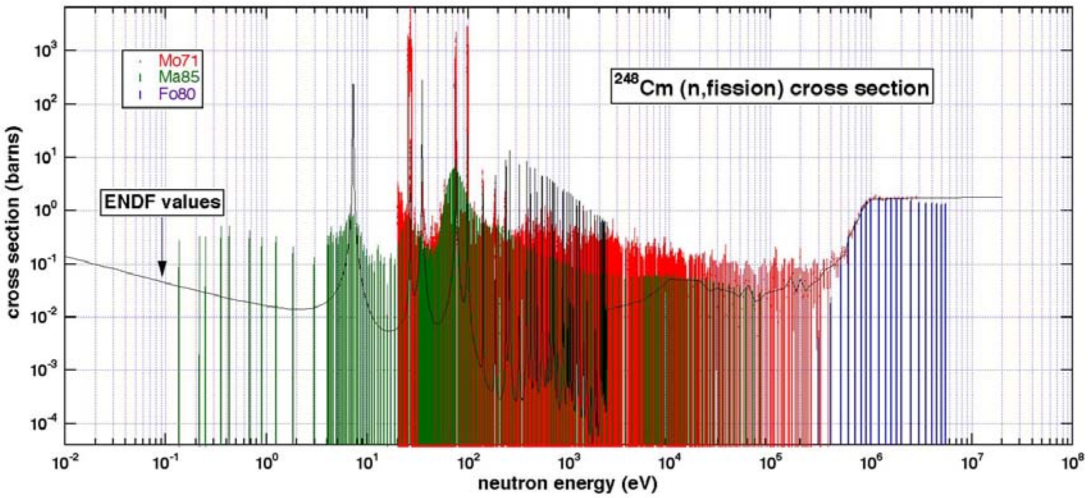
cross section (barns)
neutron energy (eV)



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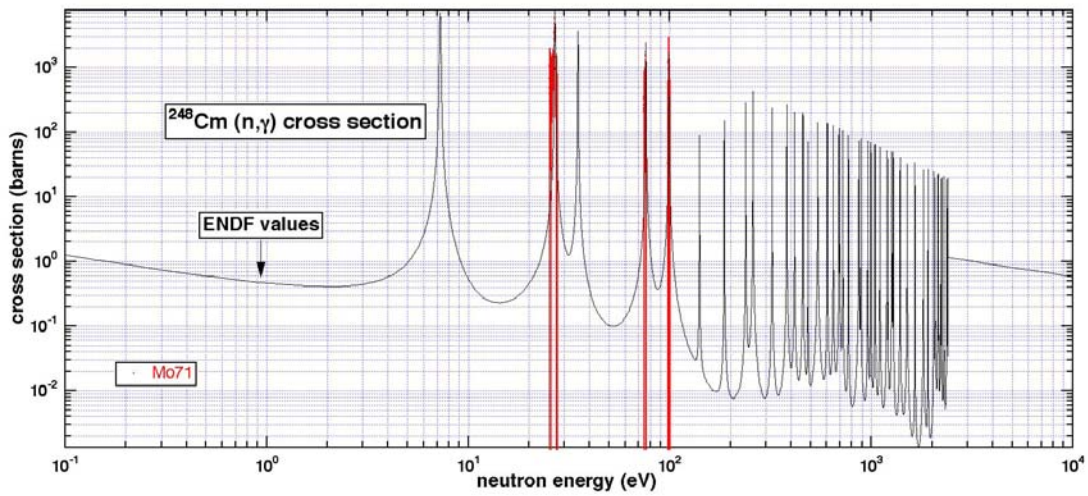
cross section (barns)
neutron energy (eV)



^{248}Cm (n,γ) cross section
ENDF values
Mo71
cross section (barns)
neutron energy (eV)

Appendix B

Multiparameter Data Collection, List-Mode Data Storage and Data Sorting

Appendix B

Multiparameter Data Collection, List-Mode Data Storage and Data Sorting

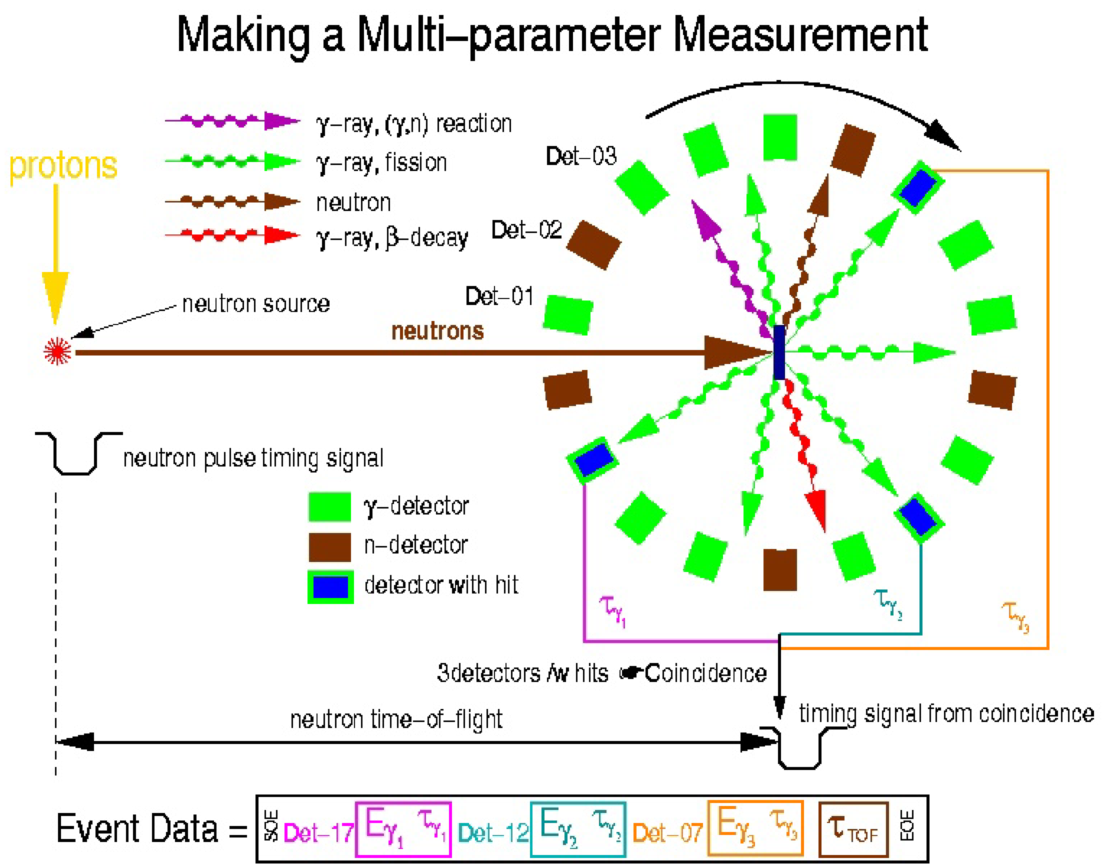


Figure B.1. Schematic representation of a multi-parameter experimental configuration and the data packet for one event.

Figure B.1 represents an experimental setup for measuring neutron interaction cross sections and other information using a pulsed neutron beam and a mixed array of radiation detectors. In this example, to simplify the case, no particle detectors are shown but when they are present, n,charged-particle cross sections, branching rations, and light fragment emission rates are included in the data. In this example, the detectors are numbered clockwise from “Det-01” just above the line representing the neutron beam to “Det-18” just below the line. Two types of detectors are shown, neutron and gamma-ray detectors. Three gamma-ray detectors are indicated as struck by radiation, and thus an “event” is deemed to have occurred and a “data packet” of the event data is collected. In this example, two or more detectors being struck by radiation is the condition that an event of interest has occurred. Detectors 7, 12, and 17 are shown to have been struck by gamma rays.

In addition to the energy ($E_{\gamma i}$) and type of radiation (γ) being recorded, the time relation ($\tau_{\gamma i}$) of the radiation striking each detector with respect to a master event signal is recorded and associated with the energy of the radiation from the i'th detector. Also, the number of the detector, and thus its position, is recorded and the time between the start of the neutron pulse leaving the neutron-generating target of the facility accelerator, and the neutron causing the event is recorded. This is the neutron time-of-flight and is directly related to the neutron energy.

With the system used in the INEEL work, multiple neutrons from each neutron pulse can be tagged so that multiple data packets from multiple events caused by different neutrons in the same neutron pulse are separately recorded. This greatly increases the amount of data that is taken during and experiment. In most prior experiments, only one neutron per pulse of neutrons could be timed or the neutrons arriving within a "time window" were observed, so that the data that could be recorded and correlated with neutron energy was lower on a per pulse or per experiment basis that with the method described here.

The system is configured such that when one detector is struck by radiation, a "time window" is opened and it is only during that time window that a signal from another detector is accepted. This time window can be from a few nanoseconds (10^{-9} seconds) to approximately 100 nanoseconds and depends on the geometry of the array and the type of radiations being detected. It is this short time window of the coincidence circuit that eliminates uncorrelated events. A system such as the one is being described here is generally configured such that the coincidence rate or "real" events is from a few hundred events per second to as high as a few thousand events per second. If the system is configured for ~10,000 events per second, a high rate for most coincidence systems, this means that on average one event occurs every 100 microseconds. With a short time window of 50 nanoseconds, the chance of two events within the same window or a random coincidence is very small. For the experiments we perform at IPNS with 12 detectors, this random rate is generally less that one count per second.

For a fission event, two (or more) fragments are formed in an excited configuration. Each unique isotope pair formed is often referred to as a fission channel. These fragments in a channel have excess energy and must promptly emit gamma rays and neutrons to come to a more stable configuration that is generally followed by beta decay. This prompt radiation is emitted by each fragment within ~10^{-12} seconds of the fission of the original nucleus, much less than the time window of the coincidence system. This means that the neutrons and gamma rays emitted by the fission process are in prompt coincidence and can be used to identify a fission event. The gamma rays that are promptly emitted by each of the fragments are unique to the fission product fragment (isotope) and can be used to identify the isotope. Since these fragments are formed at least in pairs, one gamma ray from each fragment can uniquely identify a fission event. This very process has been used to for the first time to identify the new gamma-ray spectra and level scheme of ~170 fission products.

To return to the data packet concept, many such packets are collected. It is the goal of all our experiments to collect approximately one billion (10^9) packets onto a computer medium for later analysis. This number results in sufficient statistics so that the statistical error for any one fission channel is about 0.5%. At the time of the data analysis, these data packets can be sorted into subsets of data and each subset can be analyzed independently or along with another subset. The sort criteria might well be a gamma ray of a particular energy observed in the first detector and a gamma ray of another particular energy observed in the second detector. If these two gamma-ray energies are each from a separate isotope in a particular fission channel, this sorting will result in the selection of particular fission events from large experimental data set. This subset can be used to generate a neutron spectrum for this one isotope pair channel. With the prompt fission yield of this one isotope pair channel and the incident neutron spectrum that is measured independently, the cross section for the original fissile material, as a function of the energy range of the incident neutrons, can be determined. A similar approach can be used for neutron capture interactions.

This is a powerful method that results in high accuracy, high resolution, cross section data on a fine energy binning over the energy of the spallation neutron source being used. By selecting on the multiple gamma rays associated with the neutron capture in the fissile material, the n,γ cross section can be determined from the data set of the original experiment in the same manner as the fission cross section is determined. The inelastic neutron scattering cross section can also be determined by this method as can any other cross section that emits multiple radiations.

This method allows the one large data set to be separated into smaller sets that are any one reaction channel (n,f n,γ n,n' or others) and into subsets based on the energy of the radiation of interest. The interference of one reaction channel on another channel is minimized due to the post data collection sorting process. This allows results with lower systematic errors to be obtained and the errors on the results are dominated by the statistical error of the data.

As an example, Figure B.2 is the neutron spectrum from IPNS on beamline F1 at a distance of 20 meters. This is the spectrum that is measured to determine the neutron flux. The two vertical lines are to aid in determining the region of interest. This spectrum is obtained from a continuous monitoring of the incident neutron beam by a fission chamber and neutron time-of-flight. This spectrum is also acquired on an event-by-event basis, stored in list-mode form, and sorted to match the data described below. The spectrum has been converted to neutron flux at the target location. Each bin on the x-axis represents n/cm^2-s-MeV. The function represented by this curve is $\phi(E_n)$.

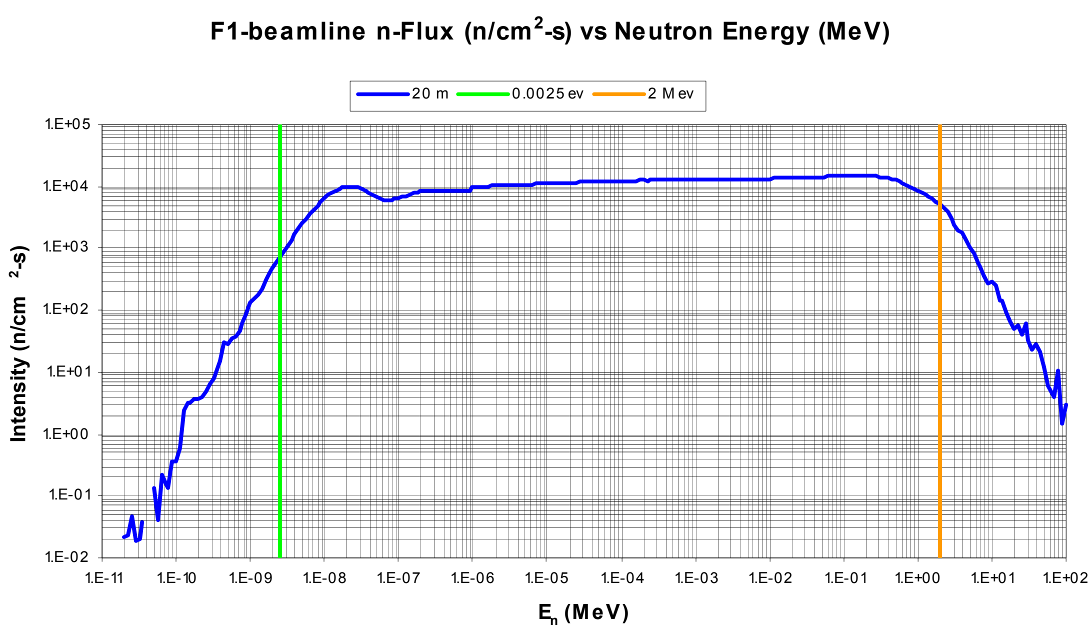


Figure B.2. Neutron-beam flux versus incident neutron energy.

Specific cross sections measurements are then made by placing a target of interest in the beam and collecting and sorting the induced emissions data as described previously. The net result of the data sorting process for a given interaction is the reaction rate per target atom as a function of incident neutron energy, $R(E_n)$, tabulated on an energy group structure determined primarily by the resolution of the time of flight measurement system. Figure B.3 illustrates a typical the result of this sorting based in selection of fission events. This process includes corrections for efficiency, binning width and all standard spectral corrections for such a measurement. This curve represents the function $R(E_n)$, that is:

$$R(E_n) = \sigma(E_n)\phi(E_n)$$

In this equation $\sigma(E_n)$ is the desired cross section and $\phi(E_n)$ is the incident neutron flux as defined above.

With these two spectra so determined, the cross section function, $\sigma(E_n) = R(E_n)/\phi(E_n)$, is the pointwise division of the two experimental spectra.

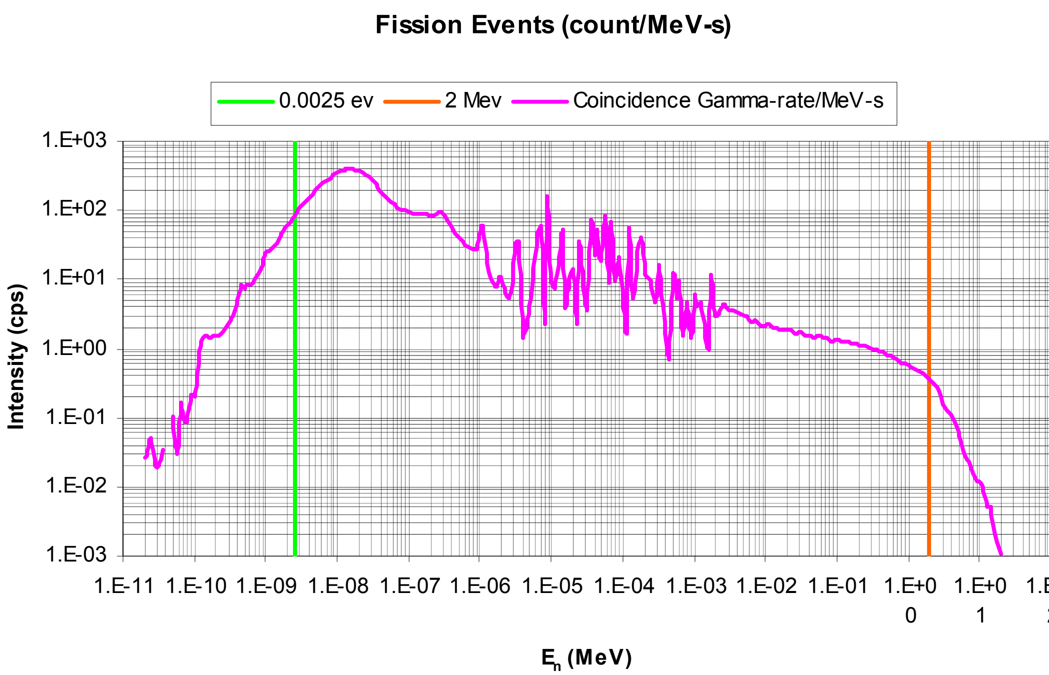


Figure B.3. Number of fission events per second versus incident neutron energy.