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NEW NEUTRON SOURCE ALGORITHMS IN THE ORIGEN-S CODE

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SUMMARY
The methods and nuclear data used to calculate the neutron source intensities and energy spectra in the ORIGEN-S code[1] have been extensively upgraded using the computational algorithms and data from the SOURCES-4B code.[2,3] The new methods allow the user to calculate multigroup neutron spectra for spontaneous fission, (α,n), and delayed neutron sources in irradiated or unirradiated materials. The (α,n) source can be calculated for any matrix of α-source and target material.

This paper describes the new methods and data used to calculate neutron source spectra and presents the results of preliminary benchmarking studies. In addition, the Windows graphical user interface (GUI) developed for ORIGEN-S and extended to support the new neutron source methods and options is described.

I. BACKGROUND
The U.S. DOE is pursuing a "dual-track" approach to the disposition of excess weapons plutonium. One approach is the immobilization of the plutonium in an inert matrix (e.g., glass or ceramic form) and subsequent disposal. The second approach involves the fabrication of mixed uranium and plutonium oxide (MOX) fuel that can be used in a commercial nuclear reactor. Safety and design evaluations of the various approaches require an accurate estimate of the neutron source. Characterization of the neutron sources for Pu-bearing materials can involve complex phenomena and require sophisticated methods.

The ORIGEN-S code is the depletion and decay analysis module of the SCALE code system developed at Oak Ridge National Laboratory and is recognized and used internationally for spent fuel characterization and source term analysis. ORIGEN-S will track the time-dependent isotopic concentrations of about 1700 nuclides, and determine aggregate radiological properties such as decay heat and neutron and gamma-ray sources and energy spectra.

The neutron source options previously implemented in ORIGEN-S provide source intensities and energy spectra for spontaneous fission neutrons, and (α,n) neutrons for either a UO_2 or a fixed borosilicate glass matrix. Although valid for many commercial spent fuel applications, the approximations in both the spontaneous fission and (α,n) neutron source algorithms limit the applicability for high plutonium bearing materials (irradiated or reprocessed fuel), MOX fuels, and alternate fuel immobilization matrix compositions.

For example, the spontaneous fission neutron spectrum calculated by ORIGEN-S was previously based on weighting the spectra for ^{244}Cm, ^{242}Cm, and/or ^{238}Pu (the dominant neutron sources in typical spent fuel). Spontaneous fission sources for other actinides were assumed to have the same energy spectra as the above nuclides. The (α,n) energy spectra were predetermined for ^{238}Pu, ^{241}Am, ^{242}Cm, and ^{244}Cm average alpha decay energies. Although the total (α,n) neutron source intensity could be calculated for other matrix compositions using the borosilicate glass option, the *energy spectrum* was always that associated with the fixed glass composition.

A recent review of the ORIGEN-S borosilicate glass spectrum using the SOURCES-4A code[4] indicated that while the total neutron production rates were in good agreement between the two codes, the energy spectra were different. This was attributed to the omission of Na and Li constituents (α,n targets) in the generation of the borosilicate glass neutron energy spectrum used in ORIGEN-S.

The assumptions used in calculating the neutron source and spectrum in ORIGEN-S may lead to inaccuracies for non-commercial spent fuel applications, particularly those involving neutron sources with a significant (α,n) component. To allow ORIGEN-S to accurately simulate a wide array of general neutron source problems, the neutron source methods were replaced with the homogeneous methods and data from the SOURCES-4B code.

II. NEUTRON SOURCE METHODS

This section summarizes the updated neutron source methods implemented in ORIGEN-S. A more detailed discussion can be found in the SOURCES-4B documentation in Refs. 2 and 3.

A. Spontaneous Fission Neutrons

Spontaneous fission neutron sources and spectra are calculated using the half-life, spontaneous fission branching fraction, number of neutrons per fission (ν), and Watt spontaneous fission spectrum parameters for 41 actinides. The half-lives used in the calculation are obtained from the ORIGEN-S ENDF/B-VI-based decay data library,[5] superceding the data in the SOURCES-4B decay library. All other data are obtained from the SOURCES-4B neutron source decay data library.[3]

The spontaneous fission neutron energy spectrum is approximated by a Watt fission spectrum using two evaluated spectral parameters, A, and B, such that

$$N(E) = Ce^{-E/A} \sinh \sqrt{BE} \qquad (1)$$

where E is the neutron energy, and C is a normalization constant.

B. (α,n) Reaction Neutrons

The (α,n) neutron source and spectra are strongly dependent on the constituent and light element content of the medium containing the α-emitting nuclides. Calculation of the neutron source requires accurate knowledge of the slowing down of the α particles and the probability of neutron production as the α particle energy decreases in the medium. The (α,n) neutron source calculation is performed assuming a homogeneous mixture in which the α-emitting nuclides are intermixed with the target nuclides. It is assumed that the dimensions of the target are much larger than the range of the α-particles; thus, all α-particles are stopped within the mixture.

The neutron yield from an alpha particle of energy E_α emitted in a homogeneous mixture with number density N, and (α,n) target nuclide density N_i, can be determined as

$$Y_{i,k} = \frac{N_i}{N} \int_0^{E_\alpha} \frac{\sigma_i(E)}{S(E)}\, dE \qquad (2)$$

where S(E) is the total stopping power, and σ(E) is the (α,n) reaction cross section for target nuclide i. The SOURCES methodology integrated in ORIGEN-S uses this expression to calculate the neutron yield from each discrete-energy alpha particle emitted by all α-emitting nuclides in the material, summed over all target nuclides. The stopping power for compounds, rather than pure elements, is approximated using the Bragg-Kleeman additivity rule. The energy-dependent elemental stopping cross sections are determined as parametric fits to data.

The values of $Y_{i,k}$ are determined in the code using a discrete numerical approximation of the integral slowing-down expression. Each alpha energy E_α is subdivided into a number of discrete energy groups, and the yield is determined for each bin using the mid-point energy of the group. The number of alpha energy groups may be set by the user. The total neutron source is determined by multiplying the total alpha source strength for each alpha energy times the respective (α,n) yield value $Y_{i,k.}$

The neutron energy spectra are calculated using nuclear reaction kinematics assuming the (α,n) reaction emits neutrons with an isotropic angular distribution in the center-of-mass system. The maximum and minimum permissible energies of the emitted neutron is determined by mass, momentum, and energy balance for each product nuclide energy level. The product nuclide levels, the product level branching data, the (α,n) reaction Q values, the excitation energy of each product nuclide level, and the branching fraction of (α,n) reactions resulting in the production of product levels are available from the nuclear data libraries. A more detailed discussion of the theory and derivation of the kinematic equations can be found in Ref. 2.

The decay data library used in the (α,n) neutron source calculation includes 89 α-emitting actinides and 7 α-emitting fission products. Neutron yield data are available for 19 target nuclides: ^{7}Li, ^{9}Be,

^{10}B, ^{11}B, ^{13}C, ^{14}N, ^{17}O, ^{18}O, ^{19}F, ^{21}Ne, ^{22}Ne, ^{23}Na, ^{25}Mg, ^{26}Mg, ^{27}Al, ^{29}Si, ^{30}Si, ^{31}P, and ^{37}Cl.

C. Delayed Neutrons

Delayed neutrons are emitted by (β^-,n) decay of short-lived fission products. The observed delay is due to the half-lives of the precursor nuclides. Delayed neutrons are only important as a neutron source for decay times of less than 10 seconds. ORIGEN-S will now calculate delayed neutron sources and spectra using an evaluated library of 105 precursor nuclides. Delayed neutron branching fractions are obtained from the SOURCES-4B decay data library. This library also contains evaluated delayed neutron spectra for each precursor nuclide. The spectra are tabulated in discrete 10 keV bins starting at 50 keV and extend up to about 2 MeV.

III. IMPLEMENTATION ISSUES

The integration of the SOURCES methods in ORIGEN-S required the addition of source options in ORIGEN-S and several modifications to the SOURCES algorithms. These include:

1. Implementation of a double precision version of the error function (ERF) was required in computing the spontaneous fission spectra to avoid small negative source values for many thermal neutron energy group structures.

2. Modifications to allowing the source calculation to proceed if an alpha energy exceeded a 6.5 MeV limit in the cross section data for some target nuclides. The code now resets the alpha energy to the maximum value allowed in the library, 6.5 MeV, and proceeds.

3. The code was modified to allow spontaneous fission and/or delayed neutron calculations to be performed even if no (α,n) source calculation is performed.

4. A source and target cutoff option was implemented in the (α,n) source calculation. When executed using the discharge compositions in a spent fuel assembly, the calculations typically include essentially all of the alpha source nuclides and targets in the data library, leading to more than 1500 source-target nuclide combinations. This can result in a significant (α,n) source computing time that is many times longer than the actual ORIGEN-S depletion calculation. The cutoff feature eliminates low-importance α-sources and target nuclides, dramatically reducing the computing time in many spent fuel problems. The alpha source removal is based on the fractional contribution of the nuclide to the total alpha decay power, and the target removal is based on the atom fraction of all target nuclides in the case. The cutoff feature is optional, and the user may still perform the neutron source calculation using all source and target nuclides if desired.

5. For oxide fuels, a significant neutron source can be produced from ^{17}O (α,n) and ^{18}O (α,n) reactions in the oxygen compounds of the fuel. ORIGEN-S provides a built-in UO_2 matrix option. If this option is invoked, the code automatically assumes the calculation is to be performed for a UO_2 fuel matrix and uses a predefined matrix composition that includes uranium and the natural isotopic distribution of ^{17}O and ^{18}O. Therefore, the oxygen isotopes are included even if they have not been explicitly defined in the user input. The α-emitting source nuclides are determined automatically from the activity of the nuclides as determined from the ORIGEN-S case. The UO_2 option is the default in ORIGEN-S.

6. A built-in borosilicate glass matrix option is also provided. The fixed matrix composition is based on a predefined borosilicate glass waste form taken from Ref. 6. The neutron yields and spectra are calculated assuming the alpha emitters are infinitely dilute in the glass matrix, i.e., fuel or other alpha-emitting constituents are not included in the matrix definition. The borosilicate glass option assumes the predefined glass matrix composition, irrespective of the actual compositions defined in the problem. The predefined composition matrix options are provided for backwards compatibility with previous versions of ORIGEN-S.

7. An arbitrary problem-specific matrix may also be specified for the (α,n) source calculation. With this option the code calculates the (α,n) neutron source and spectra based on the material compositions in the depletion or decay problem. For spent fuel applications, the code uses the nuclide concentrations after irradiation. The user may also calculate the neutron sources for a non-fuel case by

entering the initial compositions (e.g., Po-Be source) and creating a decay case. The user also has the option of modifying the matrix by blending the irradiated fuel source nuclides with a different matrix material.

8. A detailed neutron source print option was added in ORIGEN-S that prints details of both the neutron source for each source nuclide α energy and target nuclide combination, and the neutron energy spectrum for each nuclide.

IV. GRAPHICAL USER INTERFACE

The ORIGEN-S code was developed from the ORIGEN code in the late 1970s as a depletion and decay analysis module for SCALE. The focus of development was on expanding the source capabilities (i.e., addition of energy-dependent neutron spectra) and adding the ability to easily interface with other modules in SCALE depletion and shielding sequences. Since the input to ORIGEN-S is often created automatically, and the code is executed by the control sequences within SCALE, there was little motivation to improve the user input for standalone execution. As a result, ORIGEN-S has not been as widely adopted as a standalone depletion and decay code (outside of SCALE) as the ORIGEN2 code, which has offered a more user-friendly input but has more limited capabilities.

To improve the user interface with ORIGEN-S, ORNL has developed a Windows Graphical User Interface (GUI) called OrigenArp that prepares input to the code from a series of Windows menus.[7] OrigenArp also automates the preparation of problem-dependent cross sections for a burnup analysis using user-input values of the fuel enrichment, burnup, and moderator density. The cross sections are generated using the ARP module of SCALE by interpolating from a parameterized set of cross-section libraries. OrigenArp will execute ARP and ORIGEN-S, process the output to obtain specific user-specified results, and optionally display specific requested results in either tabular or graphical form.

Several enhancements were made to OrigenArp to support the new neutron source options.

- Options are provided to use the UO_2, fixed borosilicate glass matrix, or the problem-specific matrix defined by the user input for the (α,n) calculation.
- The number of α-energy bins used in the (α,n) neutron source calculation may be set.
- The fractional cutoff option was added to eliminate low-importance sources, targets, and constituents in the (α,n) calculation and reduce the calculation time.

OrigenArp executes on Windows PCs, however, the input file created by the program may be transferred to any other computing platform.

V. VALIDATION

Verification and validation of the upgraded neutron source methods and data in ORIGEN-S was performed by reproducing the homogeneous mixture sample problems used for SOURCES-4A[2] These problems involved benchmarking against the experimentally measured neutron source emission rate and spectra for a PuBe neutron source, and the neutron source rate for a UO_2F_2 solution. The results from ORIGEN-S exactly reproduce the SOURCES-4A results. The total neutron source strengths are within 17% and 10% of the measured values for the two benchmarks, respectively.

A series of additional benchmarks were used to extend the validation for this project. These benchmarks are described below.

A. Cm_2O_3 Source

Several neutron spectral measurements have been published for a ^{244}Cm source (with minor ^{242}Cm impurity) in an oxide matrix.[8,9,10] The dominant neutron source is spontaneous fission as (α,n) neutrons represent less than 1% of the total source. The measured neutron energy spectra are compared with the calculated spectrum in Fig. 1. The spontaneous fission spectrum measured by Stoddard[10] was the spectrum adopted in previous versions of ORIGEN-S for the ^{244}Cm neutron spectrum calculations. The new calculations using the Watt fission spectrum in ORIGEN-S are in good agreement with the spectra measured in Refs. 8 and 9, whereas the measurements of Stoddard appear to overestimate the low energy portion of the spectrum.

The ^{244}Cm benchmarks are particularly important to this validation study since ^{244}Cm is one of the dominant neutron sources in commercial spent fuel for cooling times from about 1 year to 100 years. Consequently, any changes in the

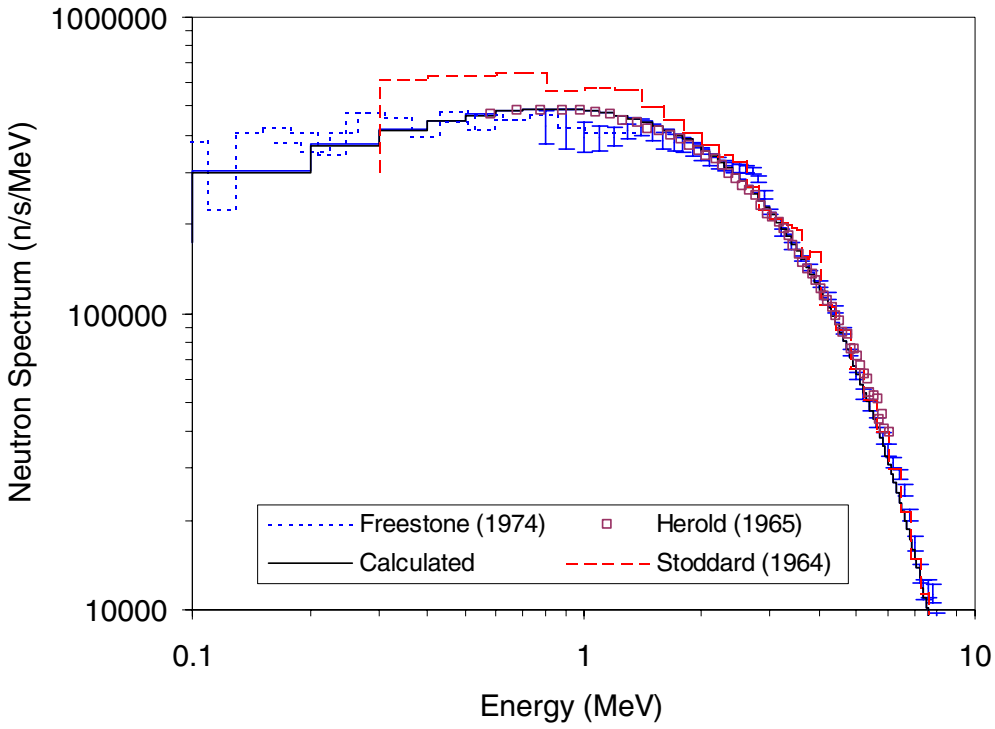

Figure 1. ^{244}Cm spontaneous fission spectrum.

^{244}Cm neutron source methods will directly effect the spent fuel neutron source.

The impact of the updated spontaneous spectra in ORIGEN-S (primarily ^{244}Cm) was evaluated for a typical concrete spent fuel storage cask. The calculations were performed for fuel with an initial enrichment of 4.5 wt % and a discharge burnup of 50 GWd/MTU. The neutron dose rates were compared over a cooling time range of 4 days to 100 years. The new source methods yielded a shielded neutron dose rate that was about 3% lower than previously calculated. Therefore, the new neutron source methods in ORIGEN-S will not significantly change previous calculations for spent commercial fuel.

B. Borosilicate Glass

The total neutron emission rates from a borosilicate glass matrix doped with a $^{238}PuO_2$ α-emitting source have been measured at the Savannah River Laboratory.[6] The glass composition in these experiments were implemented in the fixed borosilicate glass option in ORIGEN-S and were the basis of the calculated (α,n) borosilicate glass spectrum.[1] The results of four samples, each doped with 10.8 mg (sample 4), 1.03 mg (sample 5), and 0.364 mg (sample 6) of ^{238}Pu, are compared with calculated (α,n) neutron yields in Table 1.

Table 1. Borosilicate Glass Results

Glass	Neutron Emission Rate (n/s)		
Sample	Measured	Calculated	Difference
4	4,810	5,571	+16%
5	473	532	+12%
6	170	188	+11%

The uncertainty in the ^{238}Pu concentrations in the glass were about 5%. However, it was noted that the measured source may be underestimated by as much as 10% due to the calibration with a ^{252}Cf source (i.e., spectral differences). The calculated (α,n) neutron source rates are in reasonable agreement with the measurements. The predicted neutron source energy spectra from the glass are illustrated in Fig. 2, using the updated neutron

source methods and the previous methods implemented in ORIGEN-S.

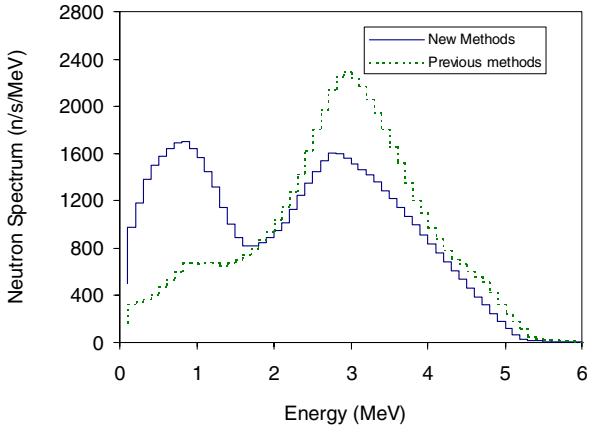

Figure 2. Predicted borosilicate glass neutron spectra using ORIGEN-S.

C. Pu-B and Pu-LiF Neutron Sources

Neutron spectral measurements have been reported[11] for mixtures of ^{238}Pu in boron and LiF target materials. PuO_2 was used in the boron source and PuF_4 was used in the LiF source. The measured and calculated spectra for Pu-B are presented in Fig. 3. The calculated spectrum is seen to be in reasonably good agreement with the measurement over the entire energy range.

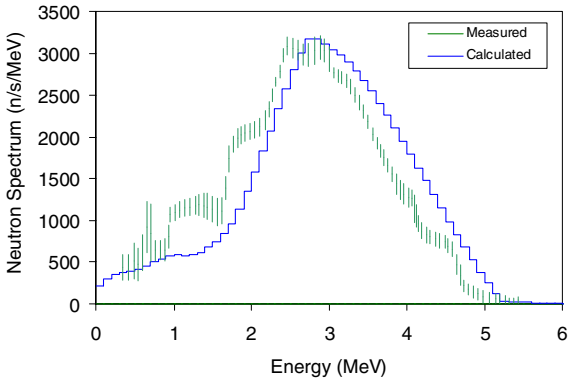

Figure 3. Pu-B neutron spectrum.

The calculated and measured spectra for the Pu-LiF source is shown in Fig. 4. The calculated spectrum is observed to be relatively poor compared to the Pu-B spectrum. This may be attributed in part to heterogeneous distribution of the Pu source and LiF target. Source particles with dimensions that are comparable to the range of the alpha particle will results in alpha particle energies in the target that are lower than those of the emitted alpha particles. That is, the slowing-down of the alpha particle within the source particle decreases the alpha particle energy as seen by the target medium. This shift in alpha particle energy can affect the neutron source and energy spectrum. Other SOURCES calculations[2] that approximate this effect using a source-target interface treatment suggest that accounting for the finite source-particle size can significantly improve the predicted spectrum.

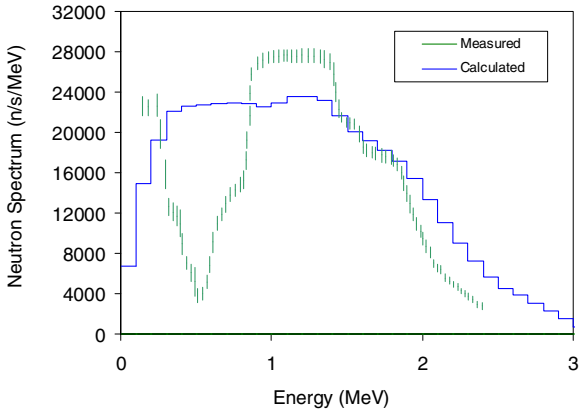

Figure 4. Pu-LiF neutron spectrum.

D. Pu-Be Neutron Source

Leakage neutron spectra from a Pu-Be source have been published in the 1.4–12.0 MeV energy range.[12] The measured spectrum is compared to the calculated source energy spectrum in Fig. 5. The calculated spectrum is in good agreement with the measurement over most of the energy range. The differences may be partly explained by the interaction of (α,n) neutrons within the source material and between the source and detector (in this case there was an empty glass sphere surrounding the source). The effect of spectral changes due to interactions of neutrons within the source has been investigated to some extent.[13]

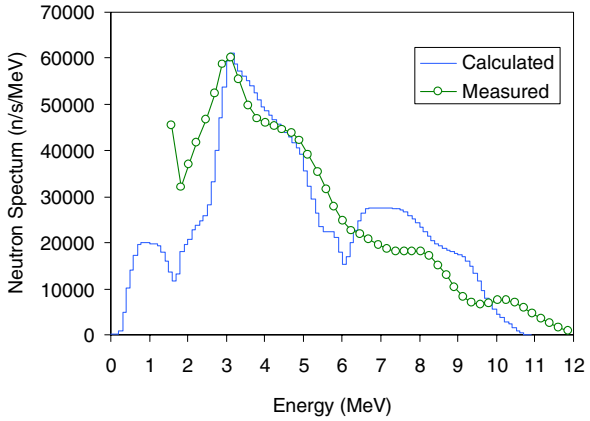

Figure 5. Pu-Be neutron spectrum.

E. Reprocessed Pu Compounds

Spectral measurements on high-exposure plutonium (< 80% ^{239}Pu) have been published for PuO_2 and PuF_4 samples.[14] The calculated spectra are compared to the measurements in Fig. 6. These experiments are poorly characterized for benchmarking purposes. The exact compositions, time since separation, etc. are not provided and have been estimated. However, the measurements do illustrate the large differences between the (α,n) spectra for different compounds, which may have a large effect on source shielding and dose rates. The shift in the spectrum for the different compounds is clearly seen in the measurements and calculations. Some of the differences between the calculated and measured spectra are probably due to interactions of (α,n) neutrons in the Pu compounds. The source size was approximately 0.9 kg in both measurements.

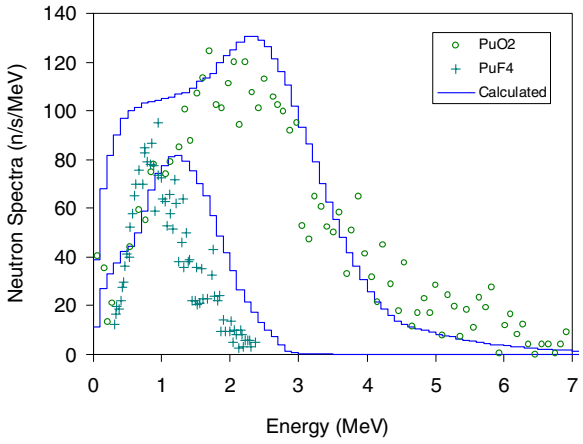

Figure 6. Neutron spectra for Pu compounds.

F. Delayed Neutron Spectra

The calculated time-integrated delayed neutron spectrum from ^{235}U thermal fission is compared to the measured spectrum[15] in Fig. 7. The results indicate that the major features of the delayed neutron spectrum are captured by the calculations. Delayed neutron spectra are available in the decay database for all of the important delayed neutron fission products (115 nuclides), and nearly all of these are available in the ORIGEN-S code.

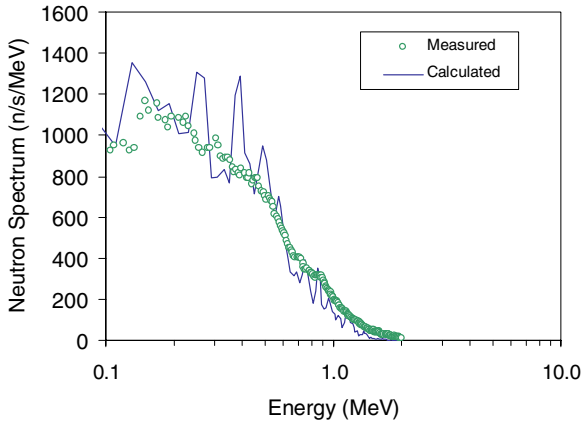

Figure 7. ^{235}U delayed neutron spectrum.

VI. CONCLUSIONS

The expanded neutron source methods in ORIGEN-S provide a powerful computational tool for the analysis of neutron emission rates and energy spectra. The neutron source methods and data, adopted from the homogeneous source methods in the SOURCES code, have been integrated with full isotopic depletion and decay capabilities provided in ORIGEN-S. The result is an updated code that can rapidly and accurately evaluate neutron emission properties for the time-dependent isotopic concentrations during decay (e.g., MOX fuel after separation) or from discharged reactor fuel or other irradiated materials.

The updated neutron source methods do not have a significant impact on commercial spent fuels because they are typically dominated by ^{244}Cm spontaneous fission, an application for which the original ORIGEN-S neutron source methods were developed.

Some of the perceived major benefits of the new source algorithms in ORIGEN-S will be in the neutron source analysis for MOX fuel. An analysis

of the neutron source emission rate for MOX fuel using typical reactor grade and weapons grade plutonium indicates that the (α,n) contribution is comparable in magnitude to the spontaneous fission neutron source for both cases. Therefore, accurate analysis of the (α,n) neutron energy spectrum is important for design and safety analysis of facilities using MOX fuel. In high-exposure PuO_2 the neutron source from (α,n) reactions on oxygen, primarily ^{18}O, may lead to neutron dose rates that are up to 50% greater than for the same amount of Pu in metal form.[16]

The new OrigenArp Windows program facilitates access to the updated ORIGEN-S neutron source capabilities in an easy-to-use, menu-driven GUI, and provide extensive data display and automated plotting capabilities.

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