APPLICATIONS OF ORIGEN TO SPENT FUEL SAFEGUARDS AND NON-PROLIFERATION

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ABSTRACT
Development of accurate and reliable methods to safeguard spent nuclear fuel residing in dry interim storage facilities is of key interest to safeguards organizations. As storage pools in many countries reach their design capacity, the number of fuel assemblies transferred from pools to dry storage is rapidly increasing worldwide. Measurements of gross neutron and gamma ray signals using the Fork detector are used routinely and widely in safeguards inspections. However, capacity to analyze and evaluate spent fuel measurement data is not very far developed. Because of the complex behavior of changes in the radiation emission signatures from different fuel types, enrichments, duration of irradiation, and decay time, several safeguards organizations are investigating increased use of modern computational methods to support the evaluation and interpretation of Fork detector results to improve the ability to verify the declared characteristics of irradiated nuclear material, and re-verify the completeness of the inventory in the future. Because these burnup codes typically track a wide range of isotopic concentrations and activities, they are also being pursued by the IAEA as a potential tool to support the analysis of environmental samples obtained from nuclear facilities worldwide. In this paper we present methods and data library development activities and several applications supporting spent fuel safeguards.

INTRODUCTION
In the United States and Europe, the number of dry storage facilities and fuel cask loading campaigns is increasing dramatically and will continue to increase over the next few years as storage pools reach their design capacities. Development of accurate and reliable methods to safeguard spent nuclear fuel residing in dry interim storage facilities is therefore of key interest to safeguards organizations. Methods are needed for verifying the characteristics of irradiated nuclear material and re-verification in the future.

Within the member states of the European Union, measurements of this type are usually performed during inspections carried out jointly between the European Commission (Euratom) and the International Atomic Energy Agency (IAEA) and are normally made using the Fork detector. The Fork detector contains dual gross-neutron and gamma-ray counting sensors. The data collected from these measurements may be used to indirectly verify the fuel type and burnup declarations made by the nuclear operator and serve as a basis for any future re-verification if required. Historically, these Fork measurements have been performed manually. However, with the sharp increase in the anticipated number of loading operations in the future, more and more measurements will be done automatically using unattended data acquisition systems. The IAEA is also investigated the implementation of spent fuel fingerprinting techniques which involve acquisition of radiation measurements, typically performed during loading operations of fuel assemblies into dry interim storage facilities, to allow indirect re-verification of the fuel in the event of a loss of continuity-of-knowledge in the future, for example, due to containment or surveillance failure.

The increasingly diverse fuel types, assembly designs, and high-burnup fuels, combined with the complexity of irradiated fuel behavior, has led safeguards organizations to investigate use of fundamentally-based modern computational methods to evaluate measurement data, replacing simple empirical relationships that are no longer valid or reliable for complex fuel cycles. The application of computational methods to simulate spent fuel isotopic inventories and neutron and gamma-ray radiation emission sources and energy spectra as a function of fuel parameters is being investigated as a means to enhance the ability to verify nuclear spent fuel. The ability to accurately predict spent fuel characteristics, and track the change in these characteristics due to decay, makes computational methods highly attractive, particularly to applications involving re-measurement of spent fuel to indirectly verify that material has not been diverted. These methods require computer codes that track the complex isotopic evolution of fuel irradiation and decay and simulate the changes in emission intensity and energy spectra with time for a wide range of potential fuel types. The IAEA also uses computational methods to predict the isotopic signatures of nuclear fuel cycle operations that can be compared with environmental sampling to verify certain declared nuclear activities. The application areas described in this paper include: (1) support for analysis of Fork detector measurements on spent fuel and (2) support for interpretation of environmental sampling data.

COMPUTATIONAL METHODS

The preeminent computer code used internationally to predict spent fuel isotopic evolution and associated radiation sources is ORIGEN (Oak Ridge Isotope GENeration). The modern version is ORIGEN-S, developed by Oak Ridge National Laboratory (ORNL) as part of the SCALE (Standardized Computer Analyses for Licensing Evaluation) code system [1]. SCALE is actively developed and maintained under a formal quality assurance (QA) configuration management plan for the U.S. Nuclear Regulatory Commission and the Department of Energy and is used for regulatory licensing and review both in the United States and internationally.

The ORIGEN code currently tracks 1119 individual fission products generated in the fuel during irradiation, 129 actinides, and 698 isotopes associated with structural and/or activation components. Most of the decay data, cross sections, and fission product yields are based on ENDF/B-VI evaluated nuclear data. Data not available from ENDF/B-VI are obtained from the Evaluated Nuclear Structure Data File (ENSDF), the Fusion Evaluated Nuclear Data Library (FENDL), and the European Activation File (EAF). One of the primary advantages of ORIGEN-S for spent fuel safeguards support applications is the ability to accurately predict the neutron and gamma ray emissions from spent fuel. The gamma ray source is generated using a recently updated database containing discrete-energy photon data for more than 1132 nuclides. This feature allows the user to generate gamma spectra in any energy-group structure of arbitrary energy resolution. The photon calculation has been validated against experiment for times less than 1 sec after fission. The neutron source calculation is based on the methods from the SOURCES code, using spontaneous fission Watt spectral parameters for 41 actinides and a matrix-dependent (alpha,n) source method using (alpha,n) cross sections and yields for alpha particles incident on 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.

The ORIGEN code has been extensively validated against destructive radiochemical measurements for more than 100 spent fuel samples from domestic and international programs involving older [2] and modern high-burnup mixed-oxide (MOX) and low-enriched uranium (LEU) fuels, decay heat measurements for more than 120 assemblies spanning cooling times up to 30 years [3], and neutron and gamma radiation measurements. The extensive validation of ORIGEN-S is a critical element in establishing the reliability of the code to support the evaluation of safeguards-relevant data.

A necessary component of accurate fuel calculations is reactor- and assembly-dependent cross-section data libraries that define the nuclear reaction rates during irradiation. The one-group cross sections used by burnup codes change as a function of irradiation (burnup), enrichment, reactor type, and operating conditions (e.g., water voiding in boiling water reactors), and depend on the fuel assembly design (fuel type and geometry configuration). It is critical to accurately characterize the cross sections for the specific fuel type being analyzed. Most reactor physics codes are not capable of analyzing the full set of fuel isotopics necessary to accurately calculate radiation sources. Those that can typically require large computing times that would make their application to routine spent fuel measurement data analysis impractical. The methods in the SCALE code system use the ORIGEN-ARP sequence to perform the rapid and accurate fuel analysis (using ORIGEN-S) that relies on interpolation (using ARP) of pre-generated cross-section libraries that cover a wide range of potential fuel types and irradiation conditions. Thus, the computing effort to generate the problem-dependent cross sections is performed in advance, allowing ORIGEN-ARP to calculate the spent fuel properties typically in several seconds, with an accuracy equal to that of detailed reactor physics calculations.

One of the significant developments in ORIGEN, to be released with SCALE 5.1 later this summer, is the significant increase in the number of reactor libraries covering more reactor types and assembly designs. In the next section we describe some of these new libraries and the methods used to generate them.

NEW REACTOR LIBRARIES

To support spent fuel analyses, ORNL has developed ORIGEN libraries for many commercial LWR and CANDU fuel designs used worldwide. Supported by DOE and in cooperation with the European Commission, European MOX, AGR, VVER, and MAGNOX fuel types have been included. ORIGEN has a fast and easy-to-use Windows graphical interface that assists users in performing fuel irradiation and decay calculations, including the interactive plotting of results such as calculated neutron and gamma-ray source spectra and nuclide concentration ratios for comparison with measurement results. Most fuel analysis calculations can be set up and executed in less than a minute.

The next release of SCALE will include many updates to the available libraries. These libraries have been generated using relatively new two-dimensional neutron transport and depletion capabilities available in the TRITON depletion sequence. TRITON uses the NEWT 2-D transport code to generate spatially dependent and material-dependent cross sections, and generate cross sections for fuel pins or assembly-average cross sections that can be used directly by ORIGEN-S for fuel inventory simulations. As an illustration of the modeling capability, the thermal flux profile for a mixed-oxide (MOX) assembly in the reactor core, surrounded by neighboring low-enriched uranium (LEU) assemblies (typical of a MOX core) is shown in Figure 1. The figure shows the large spatial

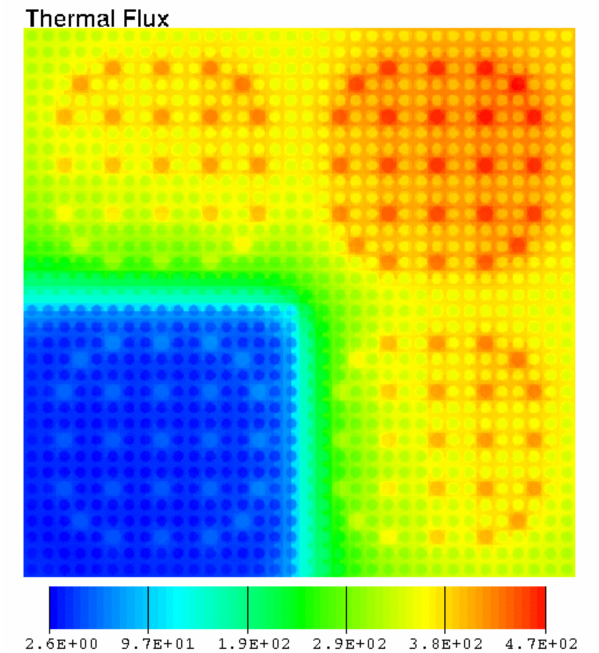


Figure 1. MOX core model with MOX assembly shown in lower-left corner, surrounded by LEU assemblies.

and energy changes in the thermal flux between the MOX and LEU assemblies, and the spectral changes within the MOX assembly, particularly near the periphery of the assembly where boundary effects become important. These effects are reflected in the ORIGEN cross-section libraries developed for the different fuel types.

This 2-D method has recently been used to update and extend the fuel libraries that can be used with ORIGEN-ARP. The libraries soon to be available are listed in Table 1. The actual geometry models for selected assembly designs are shown in Figure 2. The accuracy of these libraries for fuel analysis has been benchmarked against experiments where data are available.

APPLICATION TO UNATTENDED DATA ACQUISITION SYSTEMS

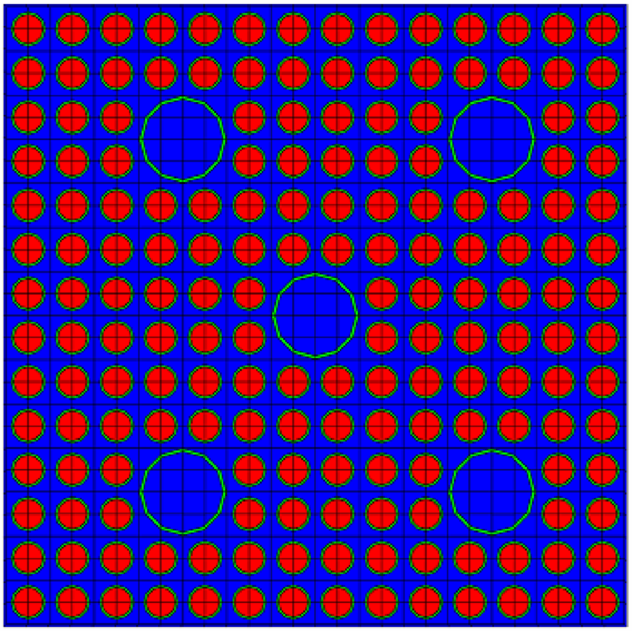
The ability to use computational methods to relate Fork measurements to declarations is being investigated by using the ORIGEN code to simulate expected neutron and gamma emission rates and energy spectra. These spectra, generated by the decay of actinide and fission product isotopes, can vary dramatically by reactor type, changes in initial fuel composition, burnup, and cooling time. Calculations have been completed and reported [4] on Fork detector simulations using MCNP for measurement configurations such as that shown in Figure 3. The results of these simulations are the energy-response curves of the detector neutron and gamma ray sensors per emitted particle in the fuel. Therefore, folding the energy response (MCNP) with the fuel emission energy spectra (ORIGEN) directly yields the counts seen by the detectors. The detector response accounts for both the change in the total source intensity as a function of burnup and time, and the change in the energy spectra of the emitted radiations. Because the computing times in ORIGEN are extremely short, the response (counts) from the detectors can be rapidly predicted over a wide range of fuel parameters, and real-time analysis of results are possible during loading operations.

Preliminary analyses using actual Fork detector measurements for MOX and LEU fuel types indicate that MCNP-ORIGEN simulations can quite accurately predict detector signals as a function of fuel burnup [4]. Repeated measurements of the same assembly over an extended decay time to validate the ability to reverify an assembly based on its source emission fingerprint has not been performed at this time. However, the decay process is considerably less complex to model than irradiation, and reliable application of this technology is anticipated.

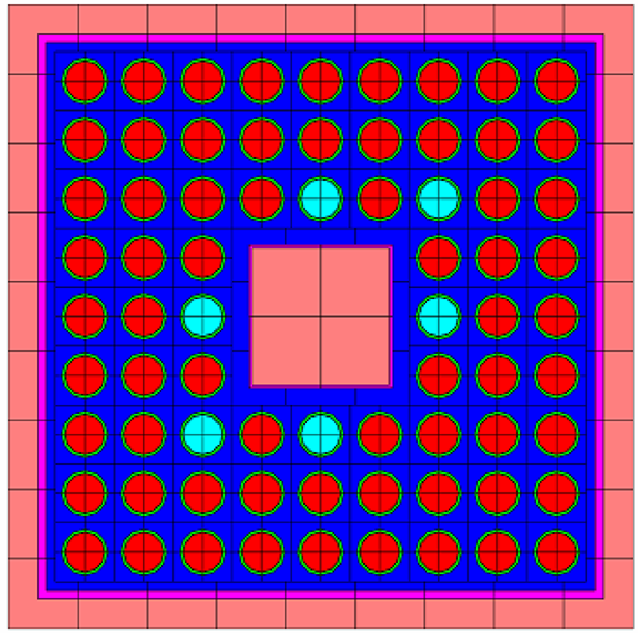
Table 1. ORIGEN Reactor Libraries to be Available in SCALE 5.1

Reactor Type*	Assembly Design Description
PWR LEU* 1.5 – 6.0 wt% < 72 GWd/t	Combustion Engineering 14x14
	Combustion Engineering 16x16
	Westinghouse 14x14
	Siemens 14x14
	Westinghouse 15x15
	Westinghouse 17x17
	Westinghouse 17x17 OFA
BWR LEU* 1.5 – 6.0 wt% < 72 GWd/t	GE 7x7
	GE 8x8
	ABB 8x8
	GE 9x9
	GE 10x10
	ATRIUM-9 9x9
	ATRIUM-10 10x10
	SVEA-64 8x8
	SVEA-100 10x10
VVER LEU*	VVER-440 1.6%, 2.4%, 3.6%
	VVER-440 3.82%
	VVER-440 4.25%
	VVER-440 4.38%
	VVER-1000
CANDU	CANDU 37 element natural uranium
	CANDU 28 element natural uranium
MAGNOX	Natural uranium
AGR	LEU
RBMK*	Under development
PWR MOX	14x14
	15x15
	16x16
	17x17
	18x18
BWR MOX	8x8-2
	9x9-1
	9x9 ATRIUM-9
	10x10 ATRIUM-10

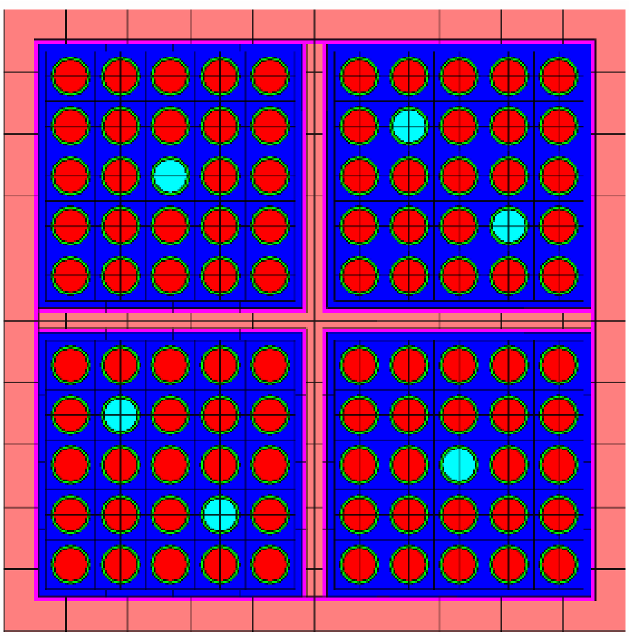
*Developed using new 2-D methods



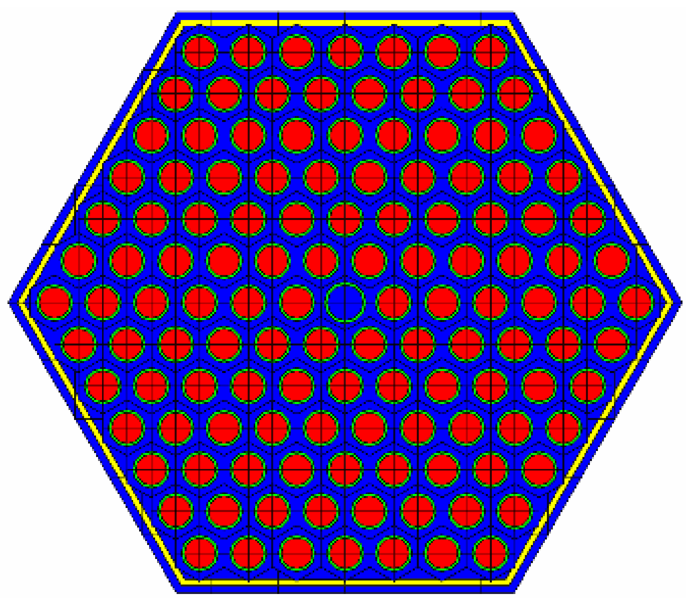
Combustion Engineering CE 14x14



ATRIUM-9



SVEA 100



VVER 400

Figure 2. Examples of selected fuel assembly models.

With a substantial increase in cask loading operations anticipated in the next few years and limited inspector resources available, more and more spent fuel measurements will likely be done automatically, using unattended data acquisition systems. A highly desirable capability of such a system is the real-time evaluation of the Fork detector signals against operator declarations and/or initial fuel data measured prior to irradiation, combined with an automated statistical analysis of the results. A requirement for use of the ORIGEN code to perform this function is an interface between the data acquisition system and the ORIGEN code. The Remote Acquisition of Data and Review (RADAR) system, developed by Euratom to standardize unattended data acquisition [5], is highly modular and is designed with the ability to easily couple with external codes. The MGA and other codes have already been integrated into the system. ORIGEN is designed as a module of the larger SCALE code system and is similarly designed to easily transfer information to and from other codes.

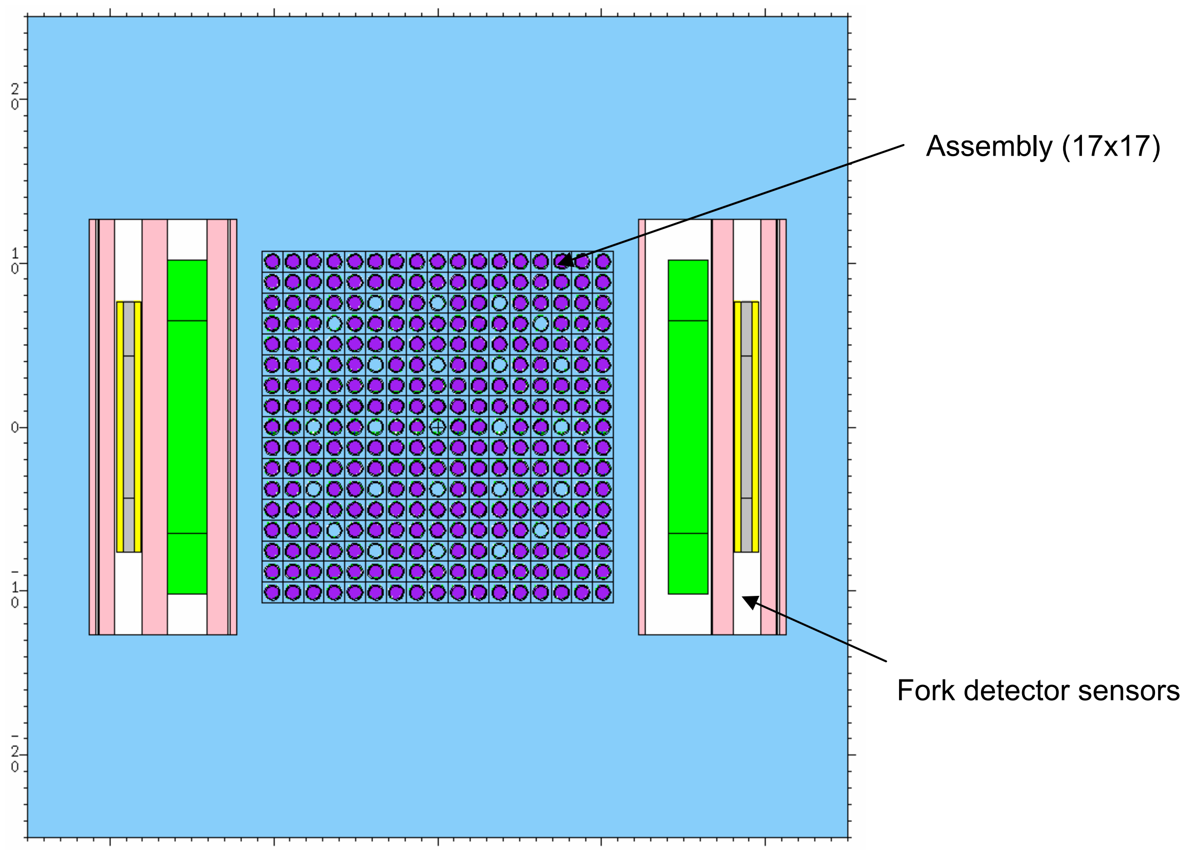


Figure 3. Instrument configuration used to develop Fork detector sensor energy response curves.

APPLICATION TO ENVIRONMENTAL SAMPLING

Destructive analysis and environmental sampling are essential safeguards tools used by the Agency. A critical component of this technology is the ability to accurately and reliably interpret measurement results to determine: (1) if the results are consistent with declared operations, (2) if deviations are consistent with measurement uncertainty and variations attributed to a range of potential operating scenarios, and (3) if there are statistically significant deviations, what types of fuels and irradiation/decay scenarios might produce the observed results? Again, the rapid execution times (several seconds) of ORIGEN, and the wide range of libraries for different reactors and fuel types now available with the code make it viable for an evaluator to rapidly analyze a wide range of potential scenarios. Also, the large number of isotopes currently tracked by ORIGEN (> 1100 fission products) can potentially expand the number of isotopes that may be used to provide important signatures of nuclear activity.

CONCLUSIONS

New reactor libraries to be distributed in SCALE 5.1 increase the number of reactor types and fuel assembly designs that can be accurately and rapidly evaluated. The Windows graphical interface developed for ORIGEN give considerable computational power to both inspectors and safeguards data evaluators to support a wide range of safeguards activities involving irradiated nuclear fuel. These methods are currently being used by safeguards organizations to support activities in the area of spent fuel verification using neutron and/or gamma measurements, and activities related to the evaluation of environmental sampling data.

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