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DEVELOPMENT OF ORIGEN-ARP METHODS AND DATA FOR LEU AND MOX SAFEGUARDS APPLICATIONS

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ABSTRACT

Many commercial nuclear power plants in Europe operate hybrid cores of low-enriched uranium (LEU) and mixed-oxide (MOX) fuel assemblies, complicating material verification and safeguards. Computational methods for nuclear fuel burnup and decay analysis provide a potentially powerful software tool for enhancing the capability and sensitivity of neutron and gamma assay techniques to verify spent nuclear fuel safeguards and discriminate between LEU and MOX fuel types. Predictive codes may be used to verify the fissile material compositions from declared records, and the predicted radiation sources may be compared with assembly measurements made using ION Fork detectors. Such analysis software can enhance verification capability compared with past practice that relied on empirical relationships to correlate the gross neutron and gamma signal with the burnup. However, in order to be of practical use to inspectors, such codes must be fast, accurate, and easy to use. Under a cooperative agreement between the U.S. Department of Energy and the European Atomic Energy Community (EURATOM), Oak Ridge National Laboratory is expanding the computational methods and data libraries in the existing ORIGEN-ARP code for the analysis of MOX and extended LEU assembly types. ORIGEN-ARP uses a unique cross-section interpolation scheme to prepare a problem-dependent library in a fast and automated manner, allowing a complete spent fuel burnup simulation to be performed in a small fraction of the time typically required by reactor codes that perform comparable tasks. The ORIGEN-ARP package has been integrated into a Windows graphically enhanced interface that requires minimal user input. Automated post-analysis data processing and plotting capabilities are provided. Calculated results obtained via these methods and data for the analysis of pressurized-water-reactor (PWR) MOX fuel have been validated against experimental data from the ARIANE International Program.

INTRODUCTION

The ION Fork detector is one of several nondestructive assay (NDA) techniques used by inspectors to safeguard spent nuclear fuel. The Fork detector consists of passive neutron fission chambers and gamma ionization chambers for concurrent neutron and gamma radiation emission assay of an assembly. Gamma and neutron verification techniques with the Fork instrument have been widely used by the European Atomic Energy Community (EURATOM) and other safeguards agencies for the verification of irradiated fuel [1]. Data analysis for such measurements was initially based upon simple empirical relationships. While simple to use, the methods are known to be nonrigorous for fuel with noncontinuous burnup, nonsymmetric axial burnup [particularly boiling-water reactor (BWR) and the Russian VVER] and variations in initial enrichment. Furthermore, the analysis provided only

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information relating to the declared burnup rather than the nuclear material concerned. This led to the conclusion that a more detailed analysis method was required. As a result, it was decided to change from the empirically based method to one that confirmed nuclear material content on the basis of measured gamma and neutron emissions being consistent with theoretical predictions.

In recent years, considerable effort has gone into improving analytical methods for data analysis using more rigorous methods that include accurate predictive codes. EURATOM has now made the decision to replace existing methods, where feasible, with validated computational methods to enhance the capability and sensitivity of Fork detector measurements. One of the principal codes selected for this purpose is ORIGEN-ARP [2,3]. Current uses by EURATOM are based solidly on the gamma and neutron Fork detector measurements of irradiated fuel. Measurements of uranium fuel assemblies have been made by EURATOM since the early 1990s. More recently, a number of measurements have included mixed-oxide (MOX) assemblies. The use of MOX fuel in commercial nuclear power reactors operated in Europe has expanded rapidly over the past decade. The European countries of France, Germany, Belgium, and Switzerland currently operate reactors that routinely utilize MOX fuel. The development of a MOX fuel analysis capability for ORIGEN-ARP, previously limited to low-enrichment-uranium (LEU) fuels, has been considered a priority area for the support of EURATOM safeguards work. In addition, LEU fuel assemblies from the Russian-designed VVER and RBMK reactors will also come under EURATOM safeguards with the future addition of central and eastern European Member States to the European Union. This paper describes the development and validation of MOX analysis methods and libraries for ORIGEN-ARP and the development of additional LEU reactor fuel libraries for EURATOM safeguards applications.

ORIGEN-ARP PACKAGE

Development of the ORIGEN-ARP methodology was initiated in the mid-1990s to effectively address the user community needs for computational speed and accuracy for a wide range of reactor conditions and assembly designs. The methodology is based on an interpolation strategy that uses the ARP module to produce cross-section libraries for the ORIGEN-S code for any enrichment, burnup, and moderator density for a specific assembly design type. The interpolation methods in ORIGEN-ARP successfully overcame limitations in ORIGEN2 methods that proved highly sensitive and led to discontinuous cross-section variations with enrichment. The ORIGEN-ARP methodology was initially released in the PC version of the SCALE 4.3 [4] code package (Standardized Computer Analyses for Licensing Evaluation). However, this version used an MS-DOS-based user interface that does not run on many current Windows-based PCs.

A fast, accurate, easy-to-use, standalone ORIGEN depletion package was still needed. In 2001, the new Windows-based graphically enhanced user interface called OrigenArp was developed. This user-friendly menu system provides ORIGEN users with tools to rapidly set up complex problems, execute the problems, and then view and interactively plot results. An important feature of the interface is a simplified input form, referred to as the Express form, which requires as few as four key input parameters (assembly type, enrichment, burnup, and power level) to perform a complete depletion analysis. In less than one minute, a user can set up and run an ORIGEN-ARP case and graphically display the results. Combining the new OrigenArp Windows interface and a new plotting program with the ORIGEN-S and ARP codes for SCALE, a standalone ORIGEN-ARP package was initially released in 2001.

The ORIGEN-ARP code system calculates nuclide concentrations for more than 1300 actinides, fission products, and activation products and determines their aggregate properties and radiation emissions.

Neutron emissions from spontaneous fission, (α,n) reactions, and delayed neutron sources, as well as gamma-ray spectra from a database of line-energy photon yields, are provided. Residual decay heat, which may be of potential future use in safeguards methods using calorimetric techniques applied to small samples, assemblies, or spent fuel pools, is also determined. The development of ORIGEN libraries for MOX fuel has not been widely pursued in the past because of the large number of different libraries that may be needed to describe the range of initial MOX fuel compositions. However, using the interpolation methods of ARP to generate libraries, it is possible to create cross sections for a wide range of conditions without excessive data storage requirements.

DEVELOPMENT OF MOX LIBRARIES

In general, the geometry, the dimensions, and the cladding material are identical for MOX and UO_2 assemblies. However, development of an interpolation strategy for MOX fuel libraries requires the consideration of many more parameters than are necessary for uranium fuels. The range of MOX material compositions, based on European experience, is summarized in Table 1. The data were compiled from records declarations for 1042 MOX fuel assemblies, including 666 PWR MOX and 376 BWR MOX assemblies. The database represents a diverse set of MOX assembly types, covering most of the designs that have been used in Europe: 14×14, 15×15, 16×16, 17×17, and 18×18 PWR assembly designs, and 8×8, 9×9, and 10×10 BWR assembly designs. Of the EURATOM member countries, only Germany is currently operating BWRs with MOX at its Gundremmingen NPP, although MOX use is foreseen in several others and one Swedish BWR (Oskarshamn).

Table 1, Composition ranges for MOX assemblies in Europe

Parameter	Minimum	Maximum
^{238}Pu/Pu wt %	0.88	2.40
^{239}Pu/Pu wt %	53.8	68.2
^{240}Pu/Pu wt %	22.3	27.3
^{241}Pu/Pu wt %	5.38	9.66
^{242}Pu/Pu wt %	2.85	7.59
^{241}Am/Pu wt %	0.71	2.59
Pu/HM[a] wt %	4.0	9.1
^{235}U/U wt %	0.24	1.18[b]
$^{239+241}$Pu/Pu wt %	65.4	73.9
fissile Pu + ^{235}U/HM wt %	3.65	5.25

[a] HM = heavy metal (U + Pu).
[b] Maximum value for PWR MOX assemblies only was 0.72 wt % ^{235}U/U.

The variable parameters incorporated into the development of MOX libraries for each assembly design included (1) total plutonium content; (2) plutonium vector — the distribution of the plutonium isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu; (3) burnup; and (4) for moderator density. The variations in the uranium isotopic vector (i.e., depleted or natural uranium) and the ^{241}Am content were found to have only a small effect on the cross sections. These parameters were therefore set to their average values for the purposes of generating cross sections.

The method developed for interpolation of cross sections for the Pu vector exploits the high correlation of the different Pu isotopes with the ^{239}Pu isotopic concentration. The correlation coefficients exceeded 0.9 for all isotopes except ^{241}Pu, based on the data for European MOX assemblies. The higher variability of ^{241}Pu is attributed to its sensitivity to variations in the MOX storage time because to its half-life of 14.4 years. An evaluation of the interpolation methods found that the overall impact of the approximations and interpolation procedure generally produced errors of less than 1% in the interpolated cross sections, which is typically less than the uncertainty in the basic cross-section evaluations. The implications for an interpolation strategy are that the Pu cross sections may be determined to reasonable accuracy using only the relative ^{239}Pu concentration as the interpolation parameter. The limitation of this method is that cross sections will not be as accurate for vectors that lie outside the range of those considered in developing the correlation relationships.

Cross-section libraries for ORIGEN-ARP were developed using the lattice physics model in the SAS2H sequence of SCALE for each of the respective assembly designs listed in Table 2. The design parameters were selected to be broadly representative of the European reactors currently operating with MOX fuel. For the 9×9 lattice, separate libraries were created for an assembly design with one small water hole (9×9 – 1) and a large water channel (9×9 – 9), similar to the Siemens ATRIUM design. The BWR assemblies that use both MOX and LEU rods (generally with gadolinium burnable poison) were modeled using typical numbers of each rod type. The cross sections were collapsed from a 238-group ENDF/B-V and –VI library into effective 1-group cross sections used by ORIGEN-S.

Table 2, MOX fuel assembly design parameters used for cross-section library production

Reactor type	Lattice type	No. of MOX rods	No. of UO_2/Gd rods	Fuel pitch (cm)	Pellet diameter (mm)	Cladding diameter (mm)	Assembly pitch (cm)
PWR	18 × 18	300	–	1.27	8.05	9.5	23.0
	17 × 17	264	–	1.26	8.19	9.5	21.5
	16 × 16	236	–	1.43	9.11	10.75	23.0
	15 × 15	205	–	1.43	9.13	10.75	21.6
	14 × 14	179	–	1.41	9.29	10.72	19.8
BWR	10 × 10 – 9	81	10	1.30	8.7	10.05	15.2
	9 × 9 – 9	60	12	1.43	9.5	10.8	15.2
	9 × 9 – 1	68	12	1.43	9.5	10.8	15.2
	8 × 8 – 2	62	14	1.63	10.3	12.3	15.2

MOX LIBRARY VALIDATION

Cross-section data and methods verification was first performed using the Phase IV-B numerical benchmark [5] developed by the Organization for Economic Cooperation and Development/Nuclear Energy Agency Nuclear Science Committee (OECD/NEANSC) Criticality Working Party, Burnup Credit Working Group, developed to address the calculation of irradiated MOX fuel compositions. The benchmark involved a series of well-defined computational problems that included first-recycle reactor-grade MOX fuel. The configuration evaluated for this work involved a typical 17×17 PWR MOX assembly lattice (Case 3). Results for the benchmark were submitted by more than six different countries, representing a wide range of computer codes and nuclear data libraries. The assembly-average results using ORIGEN-ARP were compared with the averaged values from all participants. The ORIGEN-ARP results were found to be comparable to the other independent reactor physics lattice codes and, in most cases, within the standard deviation derived from all contributed results. The time to set up and run the ORIGEN-ARP calculations using the OrigenArp user interface was typically under a minute, and, unlike most reactor physics codes, the results provided comprehensive characterization of the spent fuel properties.

Experimental validation was based on comparisons of computed actinide and fission-product isotopic concentrations in irradiated MOX fuel samples with measurements made as part of the ARIANE International Program, coordinated by Belgonucleaire, in Belgium. The nuclides selected for measurement included 17 actinides from ^{234}U to ^{249}Cm. The concentrations for a number of important fission products were also determined, including isotopes of cesium (Cs), neodymium (Nd), samarium (Sm), europium (Eu), gadolinium (Gd), and the isotopes ^{90}Sr, ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{106}Rh, ^{103}Rh, ^{109}Ag, ^{125}Sb, ^{129}I, and ^{144}Ce. MOX fuel samples were available from 14×14 assemblies irradiated in the Beznau PWR in Switzerland. Benchmark calculations were performed for three of the Beznau PWR MOX fuel samples, designated BM1, BM5, and BM6. The Beznau samples were obtained from the high-Pu-content MOX [5.5 and 6.01 wt % Pu/heavy metal (HM)] rods in the assembly. The burnup of these samples extended up to about 60 GWd/tHM. Calculations were performed using ORIGEN-ARP and HELIOS (a reactor physics lattice code developed by Studsvik Scandpower, Inc.). The calculated-to-experimental (C/E) comparisons for the nuclide concentrations for sample BM1, which were typical of all samples analyzed, are illustrated in Figure 1. Again, the results are observed to be generally consistent with the results of independent codes and with the measurements.

The isotopes ^{134}Cs and ^{137}Cs are important gamma signatures in spent fuel, and ^{244}Cm is the dominant spontaneous fission neutron source. The ability to accurately predict these isotopes is therefore particularly important for spent fuel verifications. The error in the predicted ^{244}Cm content is <7% (all samples) and in ^{137}Cs is <1%. The ^{134}Cs concentration is underpredicted consistently in most code benchmark studies, typically by 15%. This bias is attributed to probable errors in the $^{133,134}Cs$ cross-section evaluations.

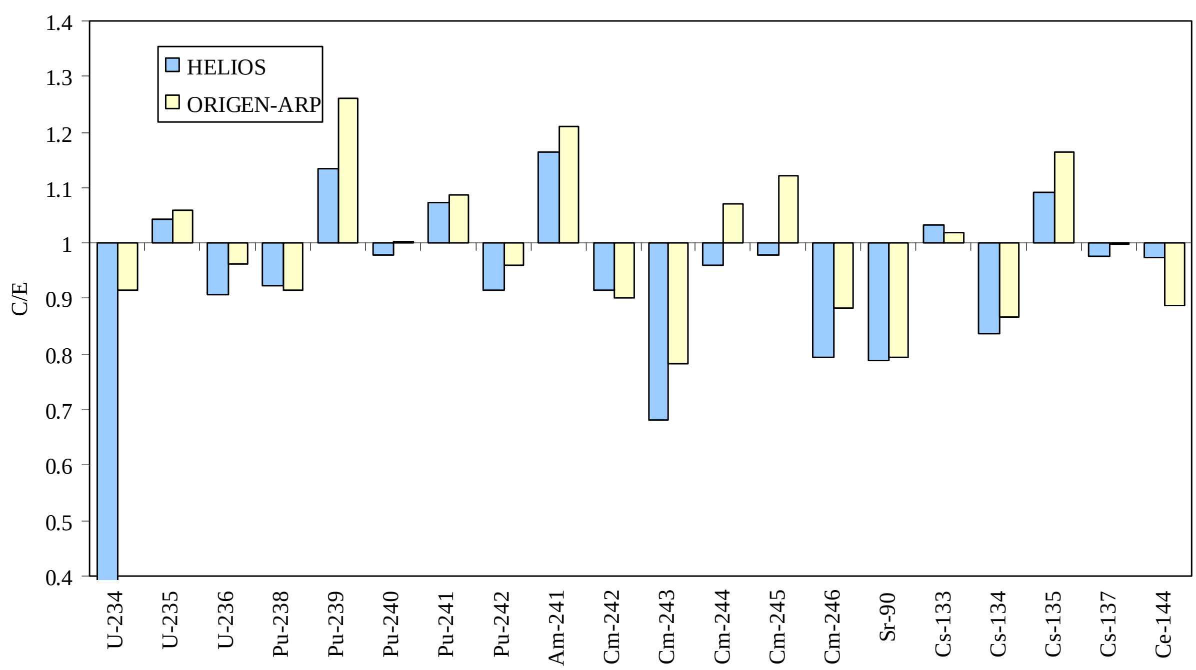

Figure 1, C/E Ratios for Selected Actinides and Fission Products in the MOX BM1 Sample

DEVELOPMENT OF ADDITIONAL URANIUM FUEL LIBRARIES

The methods and models to develop commercial LEU fuel libraries are relatively mature. Libraries for PWR 14×14, 15×15, and 17×17 assembly designs, and 8×8 BWR fuel assemblies were initially released in SCALE. Additional libraries have since been developed for 7×7 BWR assemblies and a wide range of other domestic PWR assembly types incorporating different burnable poison rod designs. To support the use of ORIGEN-ARP for unique European reactor designs, additional LEU fuel libraries are currently being developed for MAGNOX, AGR, VVER 440 and 1000, and RBMK reactor types. The development of VVER and RBMK libraries is seen as a particularly high priority for EURATOM safeguards inspectors with the future addition to the European Union of countries that operate these reactor types.

ORIGENARP WINDOWS INTERFACE FOR MOX FUEL

The new OrigenArp input form for MOX fuel has been developed and is currently being tested. The Express input form, shown in Figure 2, illustrates some of the options available to facilitate input of user specifications. The uranium, plutonium, and ^{241}Am contents may be input as either grams or as weight percent of each isotope. For input as weight percent, the code will determine the mass of each isotope from the total HM mass specified and the relative Pu content (wt % Pu/HM). The form also allows reference and loading dates to be entered. When these dates differ, the code will automatically correct the reference ^{241}Pu and ^{241}Am contents for decay and calculate concentrations representative of the actual fuel loading date.

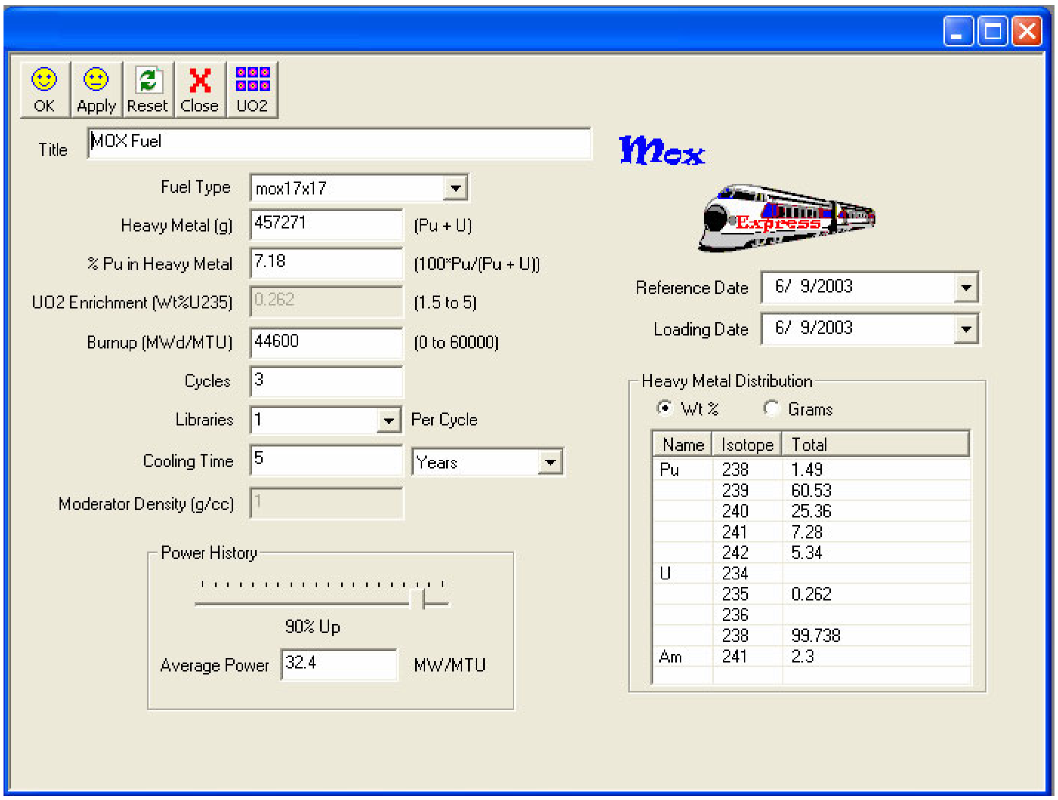

Figure 2, OrigenArp Express Form for MOX Fuel

CURRENT APPLICATION TO STRENGTHENED SAFEGUARDS

Current uses are based solidly on the gamma and neutron measurements of irradiated fuel that have been used by EURATOM for many years. Fork detector measurements on spent fuel are routinely used on assemblies being loaded into storage flasks, before they become difficult to access, in order to verify to the highest practicable level the material concerned. While the majority of spent fuel safeguards verifications require confirmation that the entire assembly contents have not been diverted, there are a number of instances in which partial defect verification is required to determine if a significant fraction of fuel rods have been diverted. In these cases the use of detailed computational methods, like ORIGEN-ARP, to improve the interpretation of Fork detector data by reducing scatter and enhancing sensitivity can improve the ability to verify discrepancies in nuclear material declaration at the partial defect level.

For each assembly being verified, the irradiation history is input to ORIGEN-ARP in order to calculate a total neutron and gamma emission, which can then be correlated to the nuclear material content. It is to be noted that while the neutron measurement alone is only indirectly related to the material content, the combination neutron and gamma data are considered to be inseparable for this safeguards verification. The verification should demonstrate consistency between the measurements and the declared fuel assembly characteristics. In recent years verifications during typical flask-loading campaigns have become very resource intensive, which has led to the development of an unattended NDA system with an associated data acquisition and analysis system. The verification and detailed analysis of assemblies with ORIGEN-ARP enables EURATOM to produce a high standard of safeguards with reduced resource requirements.

If continuity of knowledge (CoK) concerning fuel is lost, then resolving the subsequent safeguards anomaly is difficult due to access problems. One simple diversion scenario involves the substitution of core fuel with a fresh LEU assembly. Obviating this concern can be achieved only by verifying the declared fuel history and assembly contents (i.e., by confirmation of gamma and neutron emission with those predicted by ORIGEN-ARP). MOX fuel is of considerably higher strategic safeguards importance than LEU fuel and can result in a similar scenario involving the substitution of single-cycle MOX with a high burnup LEU assembly having similar neutron emissions. While core-fuel CoK problems are uncommon, such a verification approach has already been used in several European reactors to great effect.

FUTURE SAFEGUARDS APPLICATIONS

EURATOM is in the process of reevaluating its current safeguards approaches on the basis of recommendations provided by a high-level expert group [6]. The feasibility of basing an approach on a risk-based assessment methodology combined with a physical model of the nuclear fuel cycle is being assessed. An accurate prediction of nuclear material loss and production in the reactor could form a key element of such studies. One change that has already been incorporated into the approach used for on-load reactors (OLRs) involves simulation of reactor fuel evolution. ORIGEN-ARP is able to calculate the nuclear material production and loss based upon operator-declared power operation and open-source data. Assigning burnup figures to individual assemblies is beyond the scope of an independent safeguards assessment and, in any case, is unnecessary because the required assurance can be obtained by comparing integrated declared burnup values with the whole-core calculation. The OLR approach uses the reactor model as part of a material-flow-based approach rather than one based solely upon maintaining continuity of knowledge and verification.

Correlating gamma spectral data, particularly those of ^{134}Cs and ^{137}Cs, with neutron emissions has been proposed as an improved method of verifying irradiated fuel. Several instruments and analysis methods have been proposed, though none are currently accepted for safeguards purposes.

CONCLUSIONS

Computing methods based on the ORIGEN-ARP code are being adopted by EURATOM to replace many existing empirically-based methods for the interpretation of Fork detector measurements to improve nuclear materials verification. Recent development and validation of MOX fuel libraries and the development of new libraries for VVER and RBMK reactors will extend the capabilities to cover most fuel types under EURATOM safeguards.

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