ORIGEN-S Cross-Section Libraries for CANDU Used-Fuel Characterization

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ABSTRACT

A code system for producing burnup-dependent cross-section libraries for CANDU used-fuel characterization for use with the ORIGEN-S isotope generation and depletion code system is described. Benchmark results against experimental isotopic data for three CANDU-PHW reactor stations are presented. The code system couples the WIMS-AECL reactor physics analysis code with an ORIGEN-S depletion analysis to produce application-specific libraries that can be used in subsequent used-fuel analyses.

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1. INTRODUCTION

The ORIGEN series of codes are widely used within Canada and internationally for predicting the characteristics of used reactor fuel. The results from these codes are routinely applied in nuclear reactor design, radiation safety and shielding analyses, environmental safety assessments, and post-accident analyses. The accuracy of the results depends largely on the application of accurate nuclear decay data and neutron cross sections that incorporate the effects of assembly design, fuel composition, and reactor operating conditions.

The ORIGEN codes use a point-depletion model to calculate used-fuel isotopic concentrations resulting from fission and other neutron-induced transitions, decay, and fuel processing. The spatial independence of the codes requires that these effects, which affect the value of the averaged cross sections and ultimately the prediction of isotopic concentrations, be incorporated into the cross-section library prior to performing a depletion calculation. In addition, variations in the cross sections with time, caused by the changing neutron energy

spectrum and fuel isotopic concentrations, must also be taken into account. The methods used in obtaining the appropriate cross-section data, and the rigorousness of the methods, represent the primary difference between the ORIGEN series of codes.

The ORIGEN-S code [1] was recently released as part of the SCALE modular code system [2] for Standardized Computer Analyses for Licensing Evaluation. The code was designed as a depletion module of SCALE with a capability to apply multi-energy-group neutron cross sections, processed and weighted using a one-dimensional neutronic analysis of the lattice to reflect both the reactor design and time dependence of the cross sections caused by changes in the fuel isotopic concentrations. The depletion system in SCALE is designed to generate application-specific ORIGEN-S cross-section libraries from the multi-energy-group cross-section libraries in SCALE, using an automated sequence that prepares all data input and controls data interfacing between modules. Other versions of the ORIGEN code require the use of either very generic cross-section libraries, or libraries that have been pre-processed for specific reactor designs and specified fueling cycles.

The used-fuel characterization sequence in SCALE has been modified at AECL to allow a two-dimensional neutronics analysis of the reactor lattice using the WIMS-AECL [3,4] transport code. WIMS-AECL is a lattice cell code used within AECL for CANDU[1] reactor physics analyses and reactor design and development. The use of a WIMS-AECL two-dimensional analysis in place of the one-dimensional analysis of SCALE provides some significant advantages and improvements for CANDU used-fuel analyses:

- The WIMS-AECL code is the primary reactor physics code used within AECL Research for CANDU lattice calculations, and has been extensively validated for heavy-water reactor systems.

- WIMS-AECL allows a more accurate two-dimensional representation of the CANDU fuel assembly.

- The method provides access to more recent cross-section evaluations that are used with the WIMS-AECL code. This reduces the dependence on the SCALE cross-section library, which has been weighted for light-water reactor (LWR) systems, and has been shown to significantly overpredict high-energy threshold reactions when applied to CANDU analyses, and is known to exhibit biases in the prediction of uranium and plutonium reaction rates [2].

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1. CANada Deuterium Uranium, registered trademark of AECL.

- Cross-section data used in ORIGEN-S safety analyses become consistent with that used in WIMS-AECL reactor physics analyses.

This paper provides a brief description of the modified code system that couples the WIMS-AECL and ORIGEN-S codes, a summary of the benchmark comparisons carried out to date for CANDU used-fuel characterization, and planned future directions for upgrading the code system and nuclear databases.

2. DESCRIPTION OF THE ORIGEN-S CODE AND DATABASES

The ORIGEN-S code was developed as a module of the SCALE code system to calculate used-fuel isotopics and source terms for subsequent analyses. The code calculates point isotope generation and depletion resulting from neutronic transmutation, fission, radioactive decay, and fuel processing. The ORIGEN-S code is used internationally in nuclear reactor and processing plant design studies, radiation safety and shielding analyses, environmental safety assessments, and post-accident analyses, and is the reference code for CANDU used-fuel inventory characterization for the Waste Management Program within AECL. The code retains or expands the edit and processing features provided by other ORIGEN codes.

The ORIGEN-S nuclear data libraries delivered with the code contain cross-section and nuclear decay data for over 1600 isotopes. The libraries contain generic pre-weighted cross-section data based primarily on data from the original ORIGEN code [5], and fission product decay and yield data from ENDF/B-IV. Cross-section data for the isotopes ^{133}Cs, ^{242m}Am, ^{242}Am, ^{242}Cm, ^{243}Cm and ^{245}Cm, have been updated using ENDF/B-V data weighted for LWR analyses. Values of the (n,α) cross sections for many of the light elements were also corrected using ENDF/B-IV-based data. The Master Photon Libraries used to generate photon sources are based principally on data from the Evaluated Nuclear Structure Data File (ENSDF).

The primary improvement incorporated into the ORIGEN-S code is the ability to use application-specific multi-energy-group cross sections generated from a reactor physics analysis of the fuel assembly during a depletion analysis. In the coupled WIMS-AECL and ORIGEN-S code system, multigroup cross sections, weighted using the neutron energy spectrum in the fuel as calculated by WIMS-AECL, are used to create an updated nuclear database that can be input to ORIGEN-S. Multiple time-dependent libraries are used to adjust cross sections in the depletion analysis to reflect the changing fuel composition and reactor conditions. The ORIGEN-S code can also be executed as a stand-alone module independent of the coupled system using the standard nuclear database, or using updated burnup-dependent cross-section

libraries created previously with the coupled code system. Executing ORIGEN-S as a stand-alone module allows selection of fuel processing options and more detailed output descriptions than those allowed through the depletion analysis sequence.

Other improvements in the code include expanded data storage interfaces for use in subsequent calculations, use of calculated time-dependent values of recoverable energy per fission, neutron and photon source terms for any given energy group structure, and an improved neutron source calculation that includes the addition of neutrons from spontaneous fission (α,n) reactions in both UO_2 and borosilicate glass.

3. DESCRIPTION OF THE WIMS-AECL LATTICE CODE

WIMS-AECL is a deterministic multigroup transport code used for lattice cell and burnup calculations within AECL for production analyses in reactor physics design. The code is based on a version of WIMS originally obtained from the United Kingdom Atomic Energy Authority, Winfrith, and has been extensively modified. It is the primary production lattice code used for CANDU reactor physics calculations within AECL Research. Mutlidimensional reactor physics codes like WIMS-AECL generally allow full consideration of spatial effects within a lattice in a neutronics analysis.

WIMS-AECL has been validated for pin-cell lattices using cross-section data from ENDF/B-V [6]. The resonance treatment has been validated through comparisons with experiment and the results of more rigorous calculations for pin-cell and cluster fuel geometries [7]. The code has also been benchmarked for CANDU burnup calculations, with predicted inventories of the uranium and plutonium isotopes generally showing agreement of better than 1% with experimentally measured values.

The cross-section library used with WIMS-AECL is limited to isotopes of primary importance to the calculation of reactor neutronic parameters. Complete isotopic characterization, which is required for many safety-related applications, is beyond the aim and capability of specialized reactor physics like WIMS-AECL. All WIMS-AECL calculations performed in this work were run using a two-dimensional collision probability method to solve the transport equation, and an ENDF/B-V-based cross-section library.

4. MULTIGROUP CROSS-SECTION LIBRARIES

Multigroup cross sections are used in the code system to update the ORIGEN-S database with application-specific and burnup-dependent data for the depletion analysis. The code system combines two multigroup cross-section libraries: the extended

version of the SCALE 27-energy-group ENDF/B-IV-based library, and the WIMS-AECL 89-energy-group ENDF/B-V library.

The reactor physics analyses are performed using WIMS-AECL and the 89-group ENDF/B-V cross-section library developed within AECL. This library currently contains cross sections for over 150 isotopes that are generally of importance to reactor physics neutronics analyses. A total of 61 isotopes were suitable for updating the ORIGEN-S database.

The SCALE extended 27-group library is a collapsed version of the SCALE 218-group Criticality Safety Reference Library, based primarily on ENDF/B-IV, with ENDF/B-V-based data used to extend the fission product list. The library contains nuclear data for 220 individual isotopes, and was used to supplement the smaller database of the WIMS-AECL ENDF/B-V library. The 27-group library contains 13 thermal groups below 3 eV, and 5 groups above 1 MeV.

For the depletion analyses performed in this study, all isotopes available in the SCALE 27-group and WIMS-AECL 89-group library were used in updating the cross-section data in the ORIGEN-S library. Where duplicate cross-section data exist between the libraries, the code system selects the WIMS-AECL data over that from SCALE.

5. OVERVIEW OF THE CODE SYSTEM

The code system is based largely on modules from the SCALE code system, complimented with the WIMS-AECL reactor physics code for two-dimensional analysis of the reactor lattice. The interaction of the major modules in the WIMS-AECL and ORIGEN-S code system is shown in Figure 1. Each pass through the system requires new fuel compositions, and results in the creation of a single burnup-dependent library representative of the reactor assembly, along with the fuel compositions at each point in the burnup analysis.

Calculations start with the two multigroup cross-section libraries as shown at the top of Figure 1. Resonance calculations are performed on all isotopes in the SCALE library using the Nordheim Integral Treatment of NITAWL-II and initial fuel compositions. A WIMS-AECL lattice calculation is then performed to determine the neutron flux distribution in the fuel regions, and generate assembly-averaged isotopic cross sections. The WIMS-AECL resonance treatment is based on equivalence relations that relate mixtures of isotopes and heterogeneous geometries to an equivalent dilution of each isotope [7].

The two cross-section libraries are processed through the COUPLE code, which updates the standard ORIGEN-S cross-section library with the application-specific cross sections from the WIMS-AECL

and SCALE libraries. The COUPLE code collapses the multi-energy-group cross sections, using the neutron flux spectrum from WIMS-AECL, into a three-group structure used by ORIGEN-S. The 27-group cross sections are collapsed using a 27-group flux spectrum synthesized from the 89-group flux calculated by WIMS-AECL. Updating from the two data sets is performed by sequentially processing all isotopes and reaction data from the SCALE 27-group library, and substituting the 89-group data from WIMS-AECL, if it exists. If the equivalent data do not exist from WIMS-AECL, the SCALE data are retained. Since the SCALE 27-group library contains more extensive reaction data than the WIMS-AECL 89-group library, this procedure results in many isotopes containing a mix of ENDF/B-IV and -V data. Three-group flux weighting factors, THERM, RES, and FAST, are calculated in COUPLE using the WIMS-AECL flux spectrum and are applied to all remaining ORIGEN-S isotopes that are not updated with multigroup cross sections.

The updated nuclear database used by ORIGEN-S is then used to generate new fuel compositions following a short irradiation time step. A system control module subsequently reads and processes the fuel compositions after the irradiation step, writes new input interfaces for the SCALE-based codes, and writes a new material composition file for use by WIMS-AECL for the next pass through the system. The sequence proceeds until the desired irradiation is achieved. Each pass results in the creation of a time-dependent cross-section library, which can be saved for subsequent applications.

6. FUEL INVENTORY BENCHMARK STUDIES

The results of coupled WIMS-AECL and ORIGEN-S depletion analyses were benchmarked against experimental isotopic inventory data from three CANDU reactor stations: NPD, Bruce, and Pickering. The results of ORIGEN2 [8] and WIMS-AECL, run as a stand-alone code, are also included in the comparison.

ORIGEN2 is one of the codes currently used in the COG community to predict CANDU used-fuel properties. The cross-section library used with ORIGEN2 was developed at Oak Ridge National Laboratory [9] and is based on the Gentilly-2 CANDU reactor design. The cross-section data is derived from an ENDF/B-IV-based 27-group library. Adjustments were made to the ^{238}U resonance data in the ORIGEN2 library to obtain better agreement with experimental results. The ORIGEN2 code adjusts cross sections for several of the neutronically important actinides through the depletion analysis using pre-calculated data, leaving all other data constant through the analysis.

The stand-alone WIMS-AECL calculations were performed using a multi-zone representation of the fuel region, allowing burnup in

the individual regions of the assembly (fuel element rings) to proceed independently.

6.1 NPD Fuel Inventory Study

Isotopic comparisons are presented in Table 1 for uranium and plutonium inventories measured in NPD fuel [10]. The measured results for each ring of fuel elements in the 19-element bundle were weighted to produce bundle-averaged fuel compositions, the quantity compared in this study. The $^{235}U/^{238}U$ ratio was used to normalize burnup since the true burnup was not accurately known. The differences between calculation and experiment are shown for each code, and the calculated burnup is listed.

The plutonium ratios predicted by the coupled WIMS-AECL and ORIGEN-S code system are in excellent agreement with measured values. The total plutonium inventory was within 1.3% of experiment, while the $^{239}Pu/Pu$ atom ratio agreed to within 0.5%. The total plutonium inventory calculated by ORIGEN2 is overpredicted by 14%. The $^{239}Pu/Pu$ atom ratio predicted by all codes showed good agreement with experiment, while other plutonium isotopes generally showed larger differences of up to ±7%.

6.2 Bruce Fuel Inventory Study

The second set of fuel inventory measurements used in the benchmark was obtained from unpublished data from the Thorium Fuel Reprocessing Experimental program at the AECL Research Whiteshell Laboratories. Assays were available for fuel elements taken from the second, third and fourth (outer) rings of a Bruce 37-element fuel bundle (F21037C), providing characterization of 36 of the 37 elements. The experimental data were limited to uranium and plutonium inventory measurements. A detailed fuel irradiation history was available for the bundle, and was applied in all code calculations. As with the NPD fuel study, the $^{235}U/^{238}U$ ratio was used as the burnup indicator.

The bundle-averaged results are presented in Table 2. The coupled WIMS-AECL and ORIGEN-S results show excellent agreement with experiment, all quantities being within experimental error with the exception of ^{239}Pu, which showed a difference slightly larger than the measured uncertainty. The total Pu/U ratio was not available from the assay. The ORIGEN2 results for ^{239}Pu differ from experiment by 4%, while other plutonium isotopes exhibited differences from experiment of up to 20%.

6.3 Pickering Fuel Inventory Study

Actinide and fission product assays were recently performed on a single outer fuel element of Pickering-A fuel bundle 19558C in support of the AECL Waste Management Program at Whiteshell

Laboratories. The calculations assumed an outer element burnup of 222 MWh/kg initial uranium, a value derived from destructive radiochemical analyses [11]. A detailed irradiation history of the bundle was incorporated into all calculations.

While the Pickering-A fuel data are the most extensive identified to date, their use for point depletion code validation is limited since the experimental data exist for only an outer fuel element. The ORIGEN class of codes are generally designed to calculate bundle-averaged compositions. Differences between the outer element and bundle-averaged neutron flux spectrum will therefore result in errors in the averaged cross-section data. The impact was estimated, using WIMS-AECL, to be in the order of several percent for the uranium and plutonium isotopes, while the impact on the fission products is smaller. The error caused by using bundle-averaged cross sections is generally smaller than the uncertainties associated with the majority of the experimental data, with the exception of the uranium and plutonium measurements. The data presented in the benchmark comparison were therefore limited to the fission product and tranuranic isotope inventories. The WIMS-AECL results presented were calculated explicitly for the outer pin.

The results for the fission products and transuranic isotopes presented in Table 3 are generally in good agreement with experiment. The most serious discrepancy is observed in the prediction of ^{154}Eu by all codes. The difference between the WIMS-AECL result and the coupled WIMS-AECL and ORIGEN-S analysis for ^{154}Eu is due primarily to a difference in the ENDF/B-V and -IV fission product yields used in the two calculations respectively. These results suggest that errors likely exist in the ENDF/B-IV and -V fission product yields, or cross-section data for this isotope or its precursors.

The results for ^{99}Tc are presented, although problems were experienced in measuring the activity. The results for all three codes are seen to be in very good agreement. The large discrepancy between the WIMS-AECL result and the coupled WIMS-AECL and ORIGEN-S result for ^{237}Np, although within experimental uncertainty, is caused by the the spectrum difference between the calculations (outer pin and bundle-averaged, respectively). This approximation will have the greatest impact on isotopes produced principally via high-energy threshold reactions (e.g. ^{237}Np, ^{236}Pu that are produced indirectly via (n,2n) reactions).

The ORIGEN2 results for ^{244}Cm are in significantly better agreement with experiment than those of the coupled WIMS-AECL and ORIGEN-S calculations. A review of the cross-section library used in the ORIGEN2 calculations suggests the better accuracy is a result of updated 27-group cross-section data for the precursors ^{242}Cm and ^{243}Cm used in the ORIGEN2 calculations. The

ORIGEN-S libraries are currently only updated with multigroup cross sections for ^{244}Cm, a limitation in the current multigroup library. This result suggests that the current LWR weighted cross sections in ORIGEN-S are inadequate, and accurate prediction of ^{244}Cm requires updating the precursor cross sections with multigroup data processed for CANDU reactor systems. Because of the importance of ^{244}Cm as a neutron source in used fuel, the light-water spectrum-weighted cross sections for the Cm isotopes carried with the ORIGEN-S nuclear database are likely a deficiency in the present code system for application to CANDU reactor calculations.

In general, the agreement of the code predictions with experiment is good, with differences generally being of the order of the experimental uncertainty. The ORIGEN2 results showed the greatest differences from experiment, with ^{125}Sb, ^{134}Cs, ^{154}Eu and ^{155}Eu showing particularly large discrepancies.

7. CONCLUSIONS AND PLANS FOR FUTURE DEVELOPMENT

The depletion code system coupling the WIMS-AECL reactor physics code and the ORIGEN-S isotope generation and depletion code has been benchmarked against experimental measurements on CANDU reactor used-fuel isotopics, and compared with the results of ORIGEN2 and the more rigorous stand-alone calculations of WIMS-AECL. The coupled system has been shown to be in excellent agreement with experiment and with the results of WIMS-AECL calculations. The coupled code system is capable of accurately predicting used-fuel isotopics for a wide class of CANDU reactor and fuel designs.

Perhaps more importantly, the code system described is capable of generating ORIGEN-S cross-section libraries for more complex advanced-CANDU fuel designs, fuel materials, and alternate fueling cycles. The code system also provides utility modules that allow complete control over the nuclear data. The current development plans for 1992 include the following:

- The inclusion of more extensive multi-energy-group cross-section updating to correct recognized deficiencies such as the prediction of ^{244}Cm.
- Evaluation of the revised ORIGEN-S libraries containing ENDF/B-V fission product yields using the existing benchmark data, and an evaluation of recently released ENDF/B-VI yield data.
- An evaluation of revised ENDF/B-VI cross-section data for use in ORIGEN-S library updating.

- Possible expansion of the ORIGEN-S nuclear database to include ENDF/B-VI decay data that has been extensively upgraded for accurate decay heat predictions at very short and long cooling times (potential application to post-accident analysis).

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Table 1. NPD fuel inventory benchmark					
	Experiment ($\times 10^2$)		Percentage Difference from Experiment WIMS-AECL	WIMS-ORIGEN	ORIGEN2
$^{235}U/^{238}U$	0.2849	±0.3%			
Pu/U	0.313	±0.7%	+1.88	+1.25	+13.48
$^{239}Pu/Pu$	73.34	±0.1%	+0.29	+0.41	-0.95
$^{240}Pu/Pu$	22.04	±0.3%	-2.00	-1.77	+1.86
$^{241}Pu/Pu$	3.815	±0.2%	+3.75	+1.49	+6.55
$^{242}Pu/Pu$	0.812	±0.3%	+0.78	-6.56	-4.56
Calculated Burnup (MWd/MgU)			5905	5811	6042

Table 1. Isotopic Data for Uranium and Plutonium Isotopes in NPD Fuel Bundle 1016. Burnup established using $^{235}U/^{238}U$ ratio.

Table 2. Bruce fuel inventory benchmark					
	Experiment ($\times 10^2$)		Percentage Difference from Experiment WIMS-AECL	WIMS-ORIGEN	ORIGEN2
$^{235}U/^{238}U$	0.2136	±4.2%			
$^{236}U/U$	0.080	±6.3%	-2.13	-2.00	-3.50
$^{239}Pu/Pu$	65.82	±0.6%	-0.53	-0.92	+2.91
$^{240}Pu/Pu$	27.44	±2.0%	-0.44	+1.29	-4.71
$^{241}Pu/Pu$	4.96	±5.6%	+4.78	+3.00	-7.12
$^{242}Pu/Pu$	1.75	±9.7%	+8.10	+0.23	-19.57
Calculated Burnup (MWd/MgU)			8090	7834	7781

Table 2. Isotopic Data for Uranium and Plutonium Isotopes in Bruce reactor Bundle F21037C. Burnup established using $^{235}U/^{238}U$ ratio.

Table 3. Pickering activity benchmark					
Isotope	Measured Activity (Bq/kgU) (outer pin)		Percentage Difference from Experiment WIMS-AECL (outer pin)	WIMS-ORIGEN (bundle ave)	ORIGEN2 (bundle ave)
^{90}Sr	4.97+11	±4%	---	-1.81	-0.46
^{99}Tc[1]	9.35+7	±10%	+57.2	+60.6	+55.2
^{125}Sb	2.35+9	±30%	---	+20.5	+60.0
^{134}Cs	4.23+9	±5%	-4.30	-2.91	+48.2
^{137}Cs	8.42+11	±5%	---	-8.60	-6.76
^{154}Eu	8.80+9	±5%	+45.1	+88.4	+140
^{155}Eu	3.71+9	±11%	+6.33	+21.7	+49.3
^{237}Np	1.02+6	±20%	-13.8	-4.24	-4.17
^{241}Am	1.85+10	±5%	+2.05	+1.03	+6.70
^{244}Cm	7.34+8	±15%	---	+27.0	+0.91

[1] Measurement errors suspected

Table 3. Fuel inventory data for fission products and transuranics in outer element 7 of Pickering-A fuel bundle P19558C.

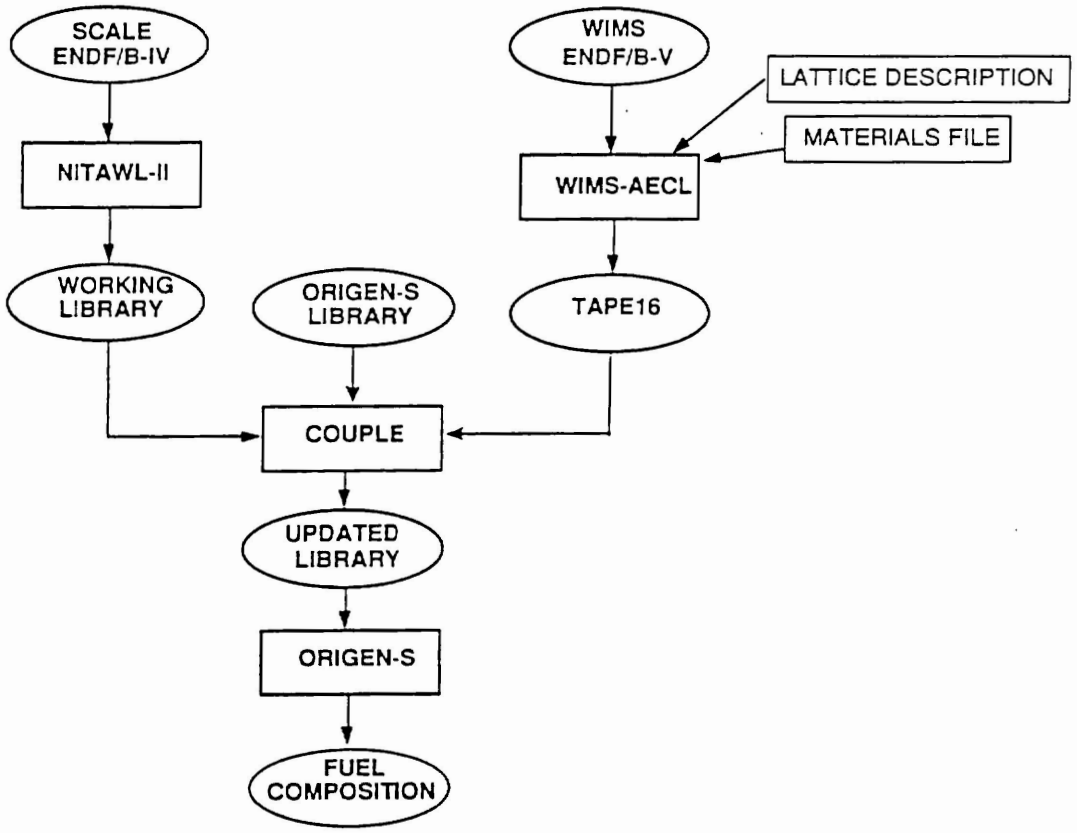

Figure 1. Overview of the coupled WIMS-AECL and ORIGEN-S code system for used-fuel characterization as used for the present calculations. Only the major code modules and data interfaces are shown.