

# Generation and validation of ORIGEN-S libraries for depletion and transmutation calculations based on JEF2.2 and EAF3 basic data

J.E. Hoogenboom <sup>a,\*</sup>, J.L. Kloosterman <sup>b</sup>

<sup>a</sup> Delft University of Technology, Interfaculty Reactor Institute, Mekelweg 15, 2629 JB Delft, Netherlands

<sup>b</sup> Netherlands Energy Research Foundation ECN, P.O. Box 1, 1755 ZG Petten, Netherlands

Received in revised form 10 July 1996

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## Abstract

The data libraries for light elements, actinides and fission products of the ORIGEN-S code for depletion and transmutation calculations in the SCALE4.1 computer code system have been updated with respect to cross-section data, radioactive-decay data and fission-product yield data using JEF2.2 as the basic data source and EAF3 as an additional source. This required the fission-product library to be extended with 201 new fission-product nuclides or isomeric states. The effect of the update of different quantities involved is evaluated with a burn-up benchmark. When ORIGEN-S is used as a stand-alone code, i.e. without regular update of cross-sections of the major nuclides due to changes in the neutron spectrum during burn-up, the results show appreciable differences in actinide and fission-product densities due to the cross-section update. The effects of updates of decay data and fission-product yields are generally small, but with noticeable exceptions. The update of fission and capture reaction energies gives a small but systematic change in actinide and fission-product concentration. The new ORIGEN-S libraries have also been converted for use with the SCALE4.2 package. © 1997 Elsevier Science S.A.

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## 1. Introduction

Present-day nuclear reactor design requires an ongoing improvement in accuracy of nuclear data. Therefore, much effort is spent to the production of up to date evaluated nuclear data libraries like ENDF/B-VI and JEF2.2. However, such basic data files are seldom used directly in reactor core calculations and the basic data are mostly pro-

cessed and converted to libraries used in actual core calculations. As the production of such libraries also requires much effort and special computer codes, users of these libraries are dependent on the timely updating of older libraries from currently available evaluated data files. This not only refers to cross-section libraries but also regards other nuclear data like fission-product yields and radioactive-decay data for fuel depletion calculations. For transmutation studies to investigate possibilities for reduction of spent fuel, these data are especially important for higher

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\* Corresponding author. Tel.: +31 15 2786962; fax: +31 15 2786422; e-mail: Hoogenboom@tudelft.iri.nl

actinides which are not of major importance for criticality calculations.

In order to base all necessary cross-section and other nuclear data needed in the calculations on the most recently available basic evaluated nuclear data files, especially the joint European file JEF2.2, the Dutch reactor physics groups at the Netherlands Energy Research Foundation ECN and at the Interfaculty Reactor Institute of the Delft University of Technology undertook a joint effort to update the data libraries for depletion and transmutation calculations used by the ORIGEN-S code as part of the SCALE system (SCALE4.1, 1992).

The ORIGEN-S code uses libraries for (three group) neutron cross sections, decay data and fission-product yields. These libraries are delivered with the SCALE-package (up to version 4.1) as three separate libraries for light elements, actinides and fission products, respectively, in card-image format. For SCALE version 4.2 the data for the three types of nuclides are combined, but a separation is made between decay data and cross-section data. These libraries are combined and converted to a binary library with the COUPLE module, which also allows updating of cross sections from fuel assembly or cell calculations. As only a limited number of nuclides will be processed in an assembly or cell calculation, for all other nuclides the cross sections from the original ORIGEN-S libraries are used in a depletion calculation as well as decay data for all nuclides and fission-product yields. Therefore, it is important to have all these data also updated to present-day standards.

This paper describes the different stages of the updating process of neutron cross sections, decay data and fission-product yields, using the JEF2.2 evaluated nuclear data file or group cross-section data derived from this basic data file. For a number of nuclides for which no cross sections are given in the JEF2.2 file, the European Activation File EAF3 (Kopecky et al., 1992) has been used. Attention is also given to nuclear data used in the ORIGEN-S code which are not in the above-mentioned ORIGEN-S libraries but included in DATA statements in the source code of ORIGEN-S.

For heating calculations the SCALE-system also provides a number of photon libraries to calculate  $\gamma$ -spectra. These libraries were not included in this updating project, although the recoverable energy as part of the decay data was updated. In this report updating from the JEF2.2 file is described. However, as the format of the JEF2 file conforms the ENDF-6 format (Rose and Dunford, 1991), as does the ENDF/B-VI file, the computer program for updating the radioactive-decay and fission-yield data can be applied to the ENDF/B-VI file as well.

## 2. Update of neutron cross-section data

The ORIGEN-S libraries contain cross-section data (and fission-product yields) for four different types of reactors, a light water reactor (LWR), a liquid metal fast breeder reactor (LMFBR), a high-temperature gas-cooled reactor (HTGR) and a molten salt breeder reactor (MSBR). The cross-section records contain thermal cross sections, resonance integrals and fission-spectrum-averaged cross sections. Also two isomer ratios (measures of the probability that nuclides are produced in a metastable state) are given in these records.

Cross sections are updated only for the LWR and LMFBR. An extensive account for the cross-section updating is given separately (Kloosterman, 1995).

### 2.1. Update from JEF2.2

Many cross sections in the ORIGEN-S data libraries have been updated with cross-section data from the JEF2.2 evaluated nuclear data file. For this purpose, the ECN cross-section data library in 172-group XMAS structure has been used. This library contains data based on JEF2.2 for about 275 actinide isotopes, light elements and fission products, and has been processed by NJOY89.62.

The nuclides in the above mentioned ECN-XMAS library have been divided in two groups: one group of nuclides for which resonance shielding has been applied (for the LWR this group consisted of 22 actinides and 32 fission products,

for the LMFBR of 22 actinides and 26 fission products), and one group for which cross sections at infinite dilution have been used (for both reactor types this group consisted of about 225 nuclides).

To represent a typical LWR the French N4 PWR design was adopted. For a representative LMFBR the French Superphénix fast reactor was taken. The first group of nuclides was used to calculate the neutron spectra of the LWR and the LMFBR as a function of burn-up. For many of these nuclides, resonance shielding calculations were done as a function of burn-up with the Bondarenko shielding method in the unresolved energy region and with the Nordheim shielding method in the resolved energy region. The burn-up calculations were done with the SCALE code system, where COUPLE was used to regularly update the cross sections in the binary ORIGEN-S data library during the burn-up sequence. The results of these calculations consist of the neutron spectra of the LWR and the LMFBR as a function of burn-up, and the three-group and one-group cross sections of these 50 nuclides (also as a function of burn-up). The above mentioned three-group cross sections for the LWR and the one-group cross sections for the LMFBR at average burn-up were converted to ORIGEN-S format by means of a conversion program.

The second group of nuclides consisted of all other nuclides and the fine-group cross sections of these nuclides were collapsed to three groups for the LWR and to one group for the LMFBR by use of the fine-group neutron fluxes in the fuel region at average burn-up of the reactor under consideration. These cross sections were also converted to ORIGEN-S format.

For the LWR, each of the three-group ORIGEN-S cross sections is relatively independent of burn-up as long as the cross-sectional changes are only due to a changing resonance-to-thermal neutron flux ratio or to a changing fast-to-thermal neutron flux ratio. This is not the case, however, for high-threshold  $(n, 2n)$  and  $(n, 3n)$  cross sections and for cross-sectional changes due to resonance shielding effects. The first two reaction types mainly depend on the neutron flux spectrum above 1 MeV; this spectrum shows a continuous

hardening as a function of burn-up, which is caused by the increasing contribution of the plutonium isotopes to the fission rate, and the resonance integrals depend on the densities of the nuclides under consideration. For the LMFBR, the one-group cross sections are relatively independent of burn-up.

## 2.2. Update from EAF3

Cross sections of nuclides which were not available from the JEF2.2 file, have been updated in the ORIGEN-S libraries with cross sections from the EAF3 file (Kopecky et al., 1992). The EAF3 file is an extensive file containing point-wise activation cross sections for 729 target nuclides with atomic numbers ranging from 1 to 96 (from Hydrogen to Curium).

For the purpose of updating the cross sections in the ORIGEN-S libraries, the cross sections in the EAF3 file have been integrated to multi-group cross sections in XMAS group structure with an LWR spectrum used as a weighting function. Then a three-group library was made for the LWR and a one-group library for the LMFBR by collapsing the multi-group library into three groups with the neutron spectrum of the PWR-N4 at average burn-up, and into one group with the neutron spectrum of the LMFBR at average burn-up. These spectra were the same as used for collapsing the JEF2.2 multi-group cross-section library into three groups for the LWR or one group for the LMFBR, as mentioned before. Finally, these few-group cross sections have been converted to ORIGEN-S format.

As mentioned before, the three card-image libraries contain two isomer ratios, which describe the production of a product in a metastable state after an  $(n, \gamma)$  reaction (FNG1) or after an  $(n, 2n)$  reaction (FN2N1). Neither ratios can be retrieved from the cross-section library based on the JEF2.2 file, because this cross-section library does not contain these fractions. Because the EAF3 file does contain cross sections for the production of a nuclide in the ground state and in the first and second metastable states, the isomer ratios could be retrieved from the EAF3 file. For this purpose, one-group cross sections for production of a nu-

clide in the ground state and in the first and second metastable states were calculated by collapsing the multi-group EAF3 library to one group by the neutron spectrum at average burn-up of the reactor under consideration (either the LWR or the LMFBR).

### 3. Update of decay data

The ORIGEN-S version 4.1 libraries contain for each nuclide or isomeric state the half life time (if it is unstable), the fraction of decay for different decay modes, the recoverable energy  $Q$  per disintegration, as well as the fraction of that energy due to  $\gamma$ - and X-rays. These data can be updated from the special purpose files with radioactive-decay data from evaluated data files like JEF and ENDF/B.

The ORIGEN-S libraries in card-image format are separated in a library for actinides and their decay products ( $Z \geq 81$ ), for fission products (including  $^3\text{H}$ ) and for 'light' elements (ranging from  $Z = 1-84$ ). This last library basically contains all the naturally occurring nuclides, their activation products from any neutron interaction and the decay products of these activated nuclei. This means that some nuclides both occur in the light-element library and the fission-product library. The JEF and ENDF/B data files do not make this distinction and list nuclides in order of their  $Z$ - and  $A$ -values, including possible isomeric states.

To update the decay data of an ORIGEN-S library a program ORSUPD was written that scans an ORIGEN-S library, reading for each unstable nuclide (or isomeric state) the decay data. Then the JEF or ENDF/B special purpose file with radioactive-decay data is searched for section  $MT = 457$  of this nuclide containing the radioactive decay data. If nuclides are found which are not present on the ORIGEN-S library, they are not added to the ORIGEN-S libraries, mainly because it cannot be simply detected whether a nuclide from JEF belongs to the light element or fission-product library. If the nuclide is found on the JEF radioactive-decay data file the record with the half live and the recoverable energy is read, as well as the record with the

modes of decay and probability. From this last record the fractions of decay for the different modes are calculated and the decay data as occurring on the ORIGEN-S library are replaced by the values derived from the JEF file and written to the updated library. The next four records for the different reactor types from the ORIGEN-S library are copied to the updated library. This process is repeated until the whole ORIGEN-S library is updated. The updating process is repeated for each ORIGEN-S library using the same JEF file. A full description of the updating program is given in a separate report (Hoogenboom and Kloosterman, 1995).

In the ORIGEN-S fission-product library there are 127 unstable nuclides, including six isomeric states, which are not present on the JEF2.2 special purpose file. No update of radioactive-decay data could be performed for these nuclides.

A few nuclides were encountered on the JEF2.2 special purpose radioactive-decay data file with zero recoverable decay energy:  $^{78}\text{Cu}$ ,  $^{93}\text{Br}$ ,  $^{94}\text{Br}$ ,  $^{101}\text{Rb}$ ,  $^{111}\text{Tc}$ ,  $^{130}\text{Cd}$ ,  $^{133}\text{In}$ ,  $^{147}\text{Xe}$  and  $^{149}\text{Ba}$ . For these nuclides, all belonging to the fission-products library of ORIGEN-S, no update of the decay energy and the fractions of different decay types is done.

### 4. Update of fission-product yields

The program ORSUPD that was used for updating decay data can also be used for updating fission-product yields. If updating of fission-product yields is requested, ORSUPD reads the JEF2.2 (or equivalent) special purpose file with direct (non-cumulative) fission-product yield data from file  $MF = 8$ , section  $MT = 454$  for all fissionable nuclides relevant to the ORIGEN-S fission-product library. These are  $^{232}\text{Th}$  (fast fission),  $^{233}\text{U}$  (thermal fission),  $^{235}\text{U}$  (thermal and fast fission),  $^{238}\text{U}$  (fast fission),  $^{239}\text{Pu}$  (thermal and fast fission),  $^{240}\text{Pu}$  (fast fission),  $^{241}\text{Pu}$  (thermal and fast fission). These nuclides are the only fissionable ones for which yields are considered in the ORIGEN-S library. For each of the reactor types (LWR, LMFBR, HTGR, MSBR) yields are specified for a set of five fissionable nuclides, but

the set of fissionable nuclides differ per reactor type as shown in Table 1 (SCALE4.1, 1992).

Although many more yield data are available on JEF2.2, the yields for only these 10 cases are used to update the ORIGEN-S fission product yield data. If JEF provides a yield value for a fission product not present in the ORIGEN-S library from one or more of the fissionable nuclides considered, this fission product is added to the updated ORIGEN-S library. This resulted in the addition of 201 new fission-product nuclides, among which 23 very light ( $Z \leq 10$ ) nuclides from ternary fission. For all these new fission products the decay data were also updated.

From the newly added nuclides,  $^{73m}\text{Zn}$  and  $^{102}\text{Rb}$  have zero decay energy on JEF2.2, in addition to the nine nuclides with zero decay energy mentioned in Section 3.

## 5. Update of other data

Besides the data provided in the ORIGEN-S libraries for depletion calculations, a calculation is also influenced by data included in the source code of ORIGEN-S. This concerns the energy released in a fission or a neutron capture. For a depletion calculation at a fixed power for a certain time interval, these energies together with the macroscopic fission cross sections determine the flux value and hence influence the results of the depletion calculation. Capture  $\gamma$ -energies are included in the ORIGEN-S code for nine light nuclides most prominent in light water reactors, for 23 fission products and for 24 actinides.

Fission energies are given for the same 24 actinides. For all other nuclides a default value of

200 MeV fission energy and 5 MeV capture energy is used. Fission and capture energies are obtained from JEF2.2 from the general purpose data file from file type MF = 3 (neutron cross sections) and section MT = 18 (total fission) and 102 (capture reaction). The value of the fission energy is the recoverable fission energy, excluding the neutrino energy. For  $^{10}\text{B}$  the energy for the (n,  $\alpha$ ) reaction was taken as the capture energy.

For most nuclides the difference of the capture energy from JEF2.2 and the original ORIGEN-S library are mostly less than 1%, with exceptions for  $^{105}\text{Rh}$  (−9.4%),  $^{134}\text{Cs}$  (+34%),  $^{241}\text{Pu}$  (−7.2%) and  $^{244}\text{Cm}$  (−14%). As these nuclides will contribute only slightly to the total capture rate, these differences cannot influence the calculation significantly. The differences in fission energy may be up to a few per cent, but for the major fissionable nuclides  $^{235}\text{U}$  and  $^{239}\text{Pu}$  they are less than 0.2%.

## 6. Further addition of nuclides to the ORIGEN-S libraries

For 103 of the nuclides (or isomeric states) newly added to the ORIGEN-S fission-product library (see Section 4) cross-section data are available in JEF2.2 or EAF3 and appropriate cross sections were derived for the LWR and LMFBR as described in Section 2 and included in the fission-product library.

A number of nuclides have (n,  $\gamma$ ) or (n, 2n) cross sections to an isomeric state, which state is not included as a nuclide on the current ORIGEN-S library. Therefore, these isomeric states are added to the updated ORIGEN-S libraries. In total 68 isomeric states are added to the light element library (18 of them with cross section for the LWR and LMFBR from EAF3), four isomeric states ( $^{207m}\text{Pb}$ ,  $^{235m}\text{U}$ ,  $^{236m}\text{Np}$ ,  $^{237m}\text{Pu}$ ) to the actinide library and two ( $^{91m}\text{Zr}$  and  $^{120m}\text{Sn}$ ) to the fission-product library. As the ground state of  $^{237}\text{Pu}$  was neither present, this nuclide was also added, as well as  $^{241}\text{Cm}$  as an important minor actinide. For the last two nuclides also cross sections were provided for the LWR and LMFBR. The decay data of the added nuclides

Table 1  
Fissionable nuclides having yield data in ORIGEN-S library

Reactor	$\gamma_1$	$\gamma_2$	$\gamma_3$	$\gamma_4$	$\gamma_5$
HTGR	$^{233}\text{U}$	$^{235}\text{U}$	$^{232}\text{Th}^a$	$^{238}\text{U}^a$	$^{239}\text{Pu}$
LWR	$^{233}\text{U}$	$^{235}\text{U}$	$^{241}\text{Pu}$	$^{238}\text{U}^a$	$^{239}\text{Pu}$
LMFBR	$^{241}\text{Pu}^a$	$^{235}\text{U}^a$	$^{240}\text{Pu}^a$	$^{238}\text{U}^a$	$^{239}\text{Pu}^a$
MSBR	$^{233}\text{U}$	$^{235}\text{U}$	$^{232}\text{Th}^a$	$^{238}\text{U}^a$	$^{239}\text{Pu}$

<sup>a</sup> Yield is for fission-spectrum energy only.

are derived from the JEF2.2 radioactive-decay data file, as far as data are given for these isomeric states.

This brings the total number of nuclides in the updated light element library to 755, for the actinide library to 107 and for the fission-product library to 1024.

## 7. Libraries for SCALE4.2

As the format of the card-image ORIGEN-S libraries of the SCALE system version 4.2 (Ryman and Hermann, 1993) was changed, a conversion program was developed to produce the appropriate format for SCALE4.2 from the updated SCALE4.1 libraries. As the SCALE4.2 format allows for delayed neutron decay, the JEF2.2 radioactive-decay data file was searched for this decay type during the conversion and the probabilities for this decay were included in the updated ORIGEN-S library. Also due to the format change the probabilities for leaving a nucleus in an isomeric state were recalculated.

It was verified that use of these libraries with ORIGEN-S 4.2 gave the same results (except for changes introduced by the delayed neutron decay) as ORIGEN-S 4.1 with the corresponding libraries.

## 8. Comparison of calculational results

To evaluate the effects of updating the various quantities involved in depletion calculations we took part I-B of the Burn-up Credit Criticality Benchmark (Brady, 1993) arranged by the Nuclear Science Committee of the OECD Nuclear Energy Agency. This concerns the depletion and transmutation of a PWR fuel rod during four operating cycles with a total burn-up of 27.35 MWd kg<sup>-1</sup> U and a 5 year cooling time. In this section the effects of the various update steps will be investigated separately. To this end the ORIGEN-S code of the SCALE system was run stand-alone, which means constant microscopic cross sections during the total life time. This was chosen in order to eliminate differences due to

cross section changes during the fuel life time. In the next section an integral test of the new ORIGEN-S library with all updated values will be performed against experimental data. Then also a regular update of microscopic cross sections for the major nuclides is taken into account.

As a reference calculation a run with the ORIGEN-S libraries as included in the SCALE4.1 code system was taken and the nuclide densities of all nuclides (101 actinides and 821 primary or secondary fission-product nuclides) saved at the end of the fourth irradiation cycle and at the end of the cooling time. Relative differences in concentrations will be shown for a number of selected fission products and most of the actinides, both at the end of the fourth irradiation period and after a 1870 days cooling time, at which time measurements were taken for the benchmark comparison. The fission products were selected either because they were requested in the benchmark comparison or because they are of interest from the neutronics point of view during operation, in case of a severe accident during operation or for the shielding of spent fuel transport.

### 8.1. Comparison of updated cross sections and SCALE4.1 libraries

The ORIGEN-S libraries were updated as far as the neutron cross sections are concerned. The cross-section updating is discussed in Section 2.

Table 2 shows the relative differences for the actinides and selected fission products. The differences for the U- and Pu-isotopes are considerable. However, one should note that normally cross sections for the most important actinides will be recalculated one or more times during an operating cycle with a new assembly or pin-cell calculation. Hence, the cross sections for these nuclides will, in general, not be taken from the ORIGEN-S libraries. The higher transuranic nuclides show large differences, up to 40%.

For the fission products there are also considerable differences. Especially the large difference for the Sm-isotopes can be noted. Other noticeable differences are: <sup>86</sup>Rb (+165%) and Cs (-36 to +9% for the various isotopes). For other fission products the difference may be a factor 2 up to a

Table 2

Concentration differences of selected fission products and actinides for ORIGEN-S/SCALE4.1 libraries with updated cross sections compared to original 4.1 libraries

Nuclide	Difference at end of irradiation (%)	Difference at end of cooling period (%)	Nuclide	Difference (%) at end of irradiation	Difference (%) at end of cooling period
<sup>3</sup> H	-0.39	-0.39	<sup>133</sup> Xe	-0.20	-0.20
<sup>79</sup> Se	-1.89	-1.89	<sup>135</sup> Xe	5.66	5.66
<sup>85</sup> Kr	2.42	2.42	<sup>133</sup> Cs	2.80	2.80
<sup>85m</sup> Kr	1.46	—	<sup>134</sup> Cs	-20.02	-20.02
<sup>87</sup> Kr	1.91	—	<sup>135</sup> Cs	8.60	8.60
<sup>88</sup> Kr	1.96	—	<sup>136</sup> Cs	-36.22	-36.22
<sup>86</sup> Rb	164.62	—	<sup>137</sup> Cs	0.05	0.05
<sup>89</sup> Sr	2.19	2.19	<sup>140</sup> Ba	0.72	0.72
<sup>90</sup> Sr	2.09	2.09	<sup>140</sup> La	0.72	0.72
<sup>91</sup> Sr	1.59	—	<sup>141</sup> Ce	-0.09	-0.09
<sup>90</sup> Y	2.17	2.09	<sup>143</sup> Ce	0.83	0.83
<sup>91</sup> Y	1.69	1.69	<sup>144</sup> Ce	1.33	1.33
<sup>95</sup> Zr	0.38	0.38	<sup>143</sup> Pr	0.89	0.89
<sup>97</sup> Zr	0.51	—	<sup>143</sup> Nd	-0.63	-0.63
<sup>95</sup> Nb	0.34	0.38	<sup>145</sup> Nd	1.30	1.30
<sup>95</sup> Mo	2.36	2.22	<sup>147</sup> Nd	-4.70	-4.70
<sup>99</sup> Mo	0.06	—	<sup>147</sup> Sm	50.97	50.97
<sup>99</sup> Tc	-1.93	-1.93	<sup>149</sup> Sm	-65.78	-65.78
<sup>99m</sup> Tc	0.06	—	<sup>150</sup> Sm	-13.49	-13.49
<sup>101</sup> Ru	-0.20	-0.20	<sup>151</sup> Sm	-72.91	-72.91
<sup>103</sup> Ru	-3.70	-3.70	<sup>152</sup> Sm	38.80	38.80
<sup>105</sup> Ru	-1.91	—	<sup>153</sup> Eu	0.29	0.29
<sup>106</sup> Ru	-2.52	-2.52	<sup>155</sup> Gd	11.46	11.46
<sup>103</sup> Rh	8.97	8.30	<sup>234</sup> U	22.73	22.73
<sup>105</sup> Rh	11.32	—	<sup>235</sup> U	-1.04	-1.04
<sup>109</sup> Ag	8.52	8.51	<sup>236</sup> U	-11.42	-11.42
<sup>126</sup> Sn	-5.43	-5.43	<sup>238</sup> U	0.08	0.08
<sup>127</sup> Sb	-5.13	—	<sup>237</sup> Np	-29.35	-29.35
<sup>129</sup> Sb	-3.05	—	<sup>239</sup> Np	-2.26	-2.26
<sup>127</sup> Te	-5.18	-5.44	<sup>238</sup> Pu	-24.09	-24.09
<sup>127m</sup> Te	-5.45	-5.44	<sup>239</sup> Pu	-13.38	-13.38
<sup>129</sup> Te	-3.41	-3.75	<sup>240</sup> Pu	9.93	9.93
<sup>129m</sup> Te	-3.75	-3.75	<sup>241</sup> Pu	27.44	27.44
<sup>131m</sup> Te	-5.20	—	<sup>242</sup> Pu	40.05	40.05
<sup>132</sup> Te	-0.86	—	<sup>241</sup> Am	39.33	39.33
<sup>131</sup> I	-1.31	—	<sup>243</sup> Am	-20.45	-20.45
<sup>132</sup> I	-1.02	—	<sup>242</sup> Cm	28.27	28.27
<sup>133</sup> I	-0.08	—	<sup>244</sup> Cm	-40.21	-40.21
<sup>135</sup> I	0.32	—	—	—	—

factor of 30. However, these fission products mostly have low densities.

### 8.2. Effects of updating decay data

The effect of updated decay data, primarily half live values, is expected to be much smaller than

for updating cross-section data. This indeed turns out to be the case for the major actinides, which show differences of less than 0.6%. For some minor actinides differences of a few per cent can be seen.

From the more important fission products most nuclides show differences of less than 0.5%. How-

Table 3  
Concentration differences of selected fission products for updated fission-product yield data

Nuclide	Difference at end of irradiation (%)	Difference at end of cooling period (%)	Nuclide	Difference at end of irradiation (%)	Difference at end of cooling period (%)
<sup>3</sup> H	-5.00	-5.00	<sup>129</sup> Te	2.05	-13.66
<sup>79</sup> Se	-1.56	-1.56	<sup>129m</sup> Te	-13.69	-13.66
<sup>85</sup> Kr	6.23	6.23	<sup>131m</sup> Te	-26.40	—
<sup>85m</sup> Kr	5.50	—	<sup>132</sup> Te	-1.77	—
<sup>87</sup> Kr	-1.17	—	<sup>131</sup> I	1.46	—
<sup>88</sup> Kr	-3.75	—	<sup>132</sup> I	-1.97	—
<sup>86</sup> Rb	11.84	11.84	<sup>133</sup> I	-0.99	—
<sup>89</sup> Sr	-1.48	-1.48	<sup>135</sup> I	-1.80	—
<sup>90</sup> Sr	-2.00	-2.00	<sup>133</sup> Xe	-1.18	—
<sup>91</sup> Sr	-1.45	—	<sup>135</sup> Xe	-2.29	—
<sup>90</sup> Y	-1.99	-2.00	<sup>133</sup> Cs	-1.11	-1.11
<sup>91</sup> Y	-1.40	-1.40	<sup>134</sup> Cs	-1.11	-1.11
<sup>95</sup> Zr	0.43	0.43	<sup>135</sup> Cs	-1.40	-1.40
<sup>97</sup> Zr	2.10	—	<sup>136</sup> Cs	11.58	—
<sup>95</sup> Nb	0.45	0.43	<sup>137</sup> Cs	-1.39	-1.39
<sup>95</sup> Mo	0.72	0.70	<sup>140</sup> Ba	-3.28	—
<sup>99</sup> Mo	0.17	—	<sup>140</sup> La	-3.24	—
<sup>99</sup> Tc	0.88	0.88	<sup>141</sup> Ce	0.24	0.24
<sup>99m</sup> Tc	0.17	—	<sup>143</sup> Ce	0.01	—
<sup>101</sup> Ru	2.82	2.82	<sup>144</sup> Ce	-0.69	-0.69
<sup>103</sup> Ru	1.69	1.69	<sup>143</sup> Pr	0.02	—
<sup>105</sup> Ru	3.50	—	<sup>143</sup> Nd	0.31	0.31
<sup>106</sup> Ru	-1.68	-1.68	<sup>145</sup> Nd	0.90	0.90
<sup>103</sup> Rh	0.55	0.60	<sup>147</sup> Nd	0.47	—
<sup>105</sup> Rh	3.50	—	<sup>147</sup> Sm	0.76	0.70
<sup>109</sup> Ag	-15.17	-15.17	<sup>149</sup> Sm	-0.14	-0.14
<sup>126</sup> Sn	24.31	24.31	<sup>150</sup> Sm	-0.39	-0.39
<sup>127</sup> Sb	9.32	—	<sup>151</sup> Sm	-1.35	-1.35
<sup>129</sup> Sb	-7.81	—	<sup>152</sup> Sm	-0.74	-0.74
<sup>127</sup> Te	9.05	7.85	<sup>153</sup> Eu	-0.85	-0.84
<sup>127m</sup> Te	7.78	7.85	<sup>155</sup> Gd	17.24	18.38

ever, from many of the other fission products differences of a factor of about two, or even more frequently occur.

### 8.3. Effects of updating fission-product yield data

The depletion results for the ORIGEN-S libraries with updated cross sections and decay data are compared with results using a fission-product library with updated yield data. This library is extended to comprise all the fission products for which yields are defined in the JEF2.2 data file. This new library has no effect on the actinides. The newly added fission products are produced only in insignificant amounts, with a

possible exception for <sup>4</sup>He (-17% to 2 · 10<sup>-4</sup> atoms kg<sup>-1</sup> UO<sub>2</sub>). The more important fission products show changes of a few per cent (see Table 3), with higher changes for <sup>86</sup>Rb, <sup>109</sup>Ag, <sup>126</sup>Sn, <sup>136</sup>Cs, <sup>155</sup>Gd and some Te-isotopes.

An approximation made in ORIGEN-S is that the fission-product yields for the fissile nuclides other than those listed in Table 1 for a certain reactor type are implicitly assumed to be zero. Including the actual yields from JEF2.2 would require a basic reprogramming of parts of the code. Therefore, it was investigated what the effect would be when yields are substituted for all fissile nuclides not having assigned fission-product yield values in the in the ORIGEN-S library.

Table 4

Comparison of nuclide concentrations or activities after 27.35 MWd kg<sup>-1</sup> UO<sub>2</sub> and 5 year cooling time with constant cross sections

Nuclide	Measured concentration <sup>a</sup> or activity	Calculated concentration <sup>a</sup> or activity	C/E value updated library	C/E value SCALE4.1 library
<sup>234</sup> U	1.60 10 <sup>-1</sup>	1.611 10 <sup>-1</sup>	1.01	0.83
<sup>235</sup> U	8.47 10 <sup>0</sup>	7.978 10 <sup>0</sup>	0.94	0.95
<sup>236</sup> U	3.14 10 <sup>0</sup>	3.141 10 <sup>0</sup>	1.00	1.13
<sup>238</sup> U	8.425 10 <sup>2</sup>	8.381 10 <sup>2</sup>	0.99	0.99
<sup>237</sup> Np	1.89 10 <sup>-4*</sup>	1.607 10 <sup>-4</sup>	0.85	1.20
<sup>238</sup> Pu	1.012 10 <sup>-1</sup>	7.848 10 <sup>-2</sup>	0.78	0.96
<sup>239</sup> Pu	4.264 10 <sup>0</sup>	3.672 10 <sup>0</sup>	0.86	0.99
<sup>240</sup> Pu	1.719 10 <sup>0</sup>	1.858 10 <sup>0</sup>	1.08	0.98
<sup>241</sup> Pu	6.812 10 <sup>-1</sup>	6.740 10 <sup>0</sup>	0.99	0.78
<sup>242</sup> Pu	2.886 10 <sup>-1</sup>	2.818 10 <sup>-1</sup>	0.98	0.69
<sup>241</sup> Am	8.56 10 <sup>-1*</sup>	7.989 10 <sup>-1*</sup>	0.93	0.70
<sup>79</sup> Se	4.55 10 <sup>-5*</sup>	2.460 10 <sup>-4*</sup>	5.41	5.67
<sup>90</sup> Sr	4.59 10 <sup>1*</sup>	4.829 10 <sup>1*</sup>	1.05	1.07
<sup>99</sup> Tc	9.59 10 <sup>-3*</sup>	1.056 10 <sup>-2*</sup>	1.10	1.12
<sup>126</sup> Sn	1.25 10 <sup>-4*</sup>	5.478 10 <sup>-4*</sup>	4.38	3.71
<sup>135</sup> Cs	4.16 10 <sup>-4*</sup>	4.232 10 <sup>-4*</sup>	1.02	0.95
<sup>137</sup> Cs	6.71 10 <sup>1*</sup>	6.715 10 <sup>1*</sup>	1.00	1.04

<sup>a</sup> Concentrations in mg g<sup>-1</sup> UO<sub>2</sub>.\*, Denotes activity values; expression in mCi g<sup>-1</sup> UO<sub>2</sub>.

Although it is not straightforward which nuclide should be selected as a substitute, for practical reasons <sup>239</sup>Pu was chosen. This required only small modifications to the ORIGEN-S code. The differences for the fission products turned out to be small, up to a few tenth of a percent and always resulting in an increase in atom densities, as can be expected. These differences will increase when the burn-up of the fuel increases or when MOX fuel is used.

#### 8.4. Effects of updating reaction Q-values

To see the effect of updating fission and capture energy values for the most important actinides and fission products a run with a special version of ORIGEN-S with the updated Q-values was made. From inspection of the ORIGEN-S output one can see the differences in total energy released per fission and the differences in flux level to obtain the same power. The total energy per fission is decreased by 0.37% and the initial flux level increased by this percentage. At the end of the irradiation period the flux level is increased by 0.52%. The effects on actinides and fission prod-

ucts are correspondingly small. However, the flux increase affects all nuclides systematically. The concentration of <sup>235</sup>U is 0.5% less. The higher actinides concentrations increase up to 2%.

### 9. Integral validation of libraries

In Section 8 only differences are shown after update of certain types of data relevant for burn-up calculations. In order to validate the fully updated ORIGEN-S libraries the concentrations (or activity) of specific nuclides can be compared with measured data from the benchmark test after the 5 year cooling time (Brady, 1993). Table 4 shows the results of an ORIGEN-S run with the fully updated library and code data for those nuclides for which experimental data are available. Note that the same cross sections are used for each irradiation period. For comparison the ratio of calculated to experimental values using the original SCALE4.1 card-image libraries with the same spectral parameters THERM, RES and FAST (0.54, 0.2516 and 2.005, respectively), is also shown. Except for the <sup>238</sup>Pu and <sup>239</sup>Pu con-

Table 5

Comparison of nuclide concentrations or activities after 27.35 MWd kg<sup>-1</sup> UO<sub>2</sub> and 5 year cooling time with regularly updated cross sections

Nuclide	Measured concentration <sup>a</sup> or activity	Calculated concentration <sup>a</sup> or activity	C/E value SAS6 run
<sup>234</sup> U	160 10 <sup>-1</sup>	1.587 10 <sup>-1</sup>	0.99
<sup>235</sup> U	8.47 10 <sup>0</sup>	8.448 10 <sup>0</sup>	1.00
<sup>236</sup> U	3.14 10 <sup>0</sup>	3.103 10 <sup>0</sup>	0.99
<sup>238</sup> U	8.425 10 <sup>2</sup>	8.366 10 <sup>2</sup>	0.99
<sup>237</sup> Np	1.89 10 <sup>-4*</sup>	1.936 10 <sup>-4</sup>	1.02
<sup>238</sup> Pu	1.012 10 <sup>-1</sup>	9.259 10 <sup>-2</sup>	0.91
<sup>239</sup> Pu	4.264 10 <sup>0</sup>	4.706 10 <sup>0</sup>	1.10
<sup>240</sup> Pu	1.719 10 <sup>0</sup>	1.768 10 <sup>0</sup>	1.03
<sup>241</sup> Pu	6.812 10 <sup>-1</sup>	7.185 10 <sup>0</sup>	1.05
<sup>242</sup> Pu	2.886 10 <sup>-1</sup>	2.841 10 <sup>-1</sup>	0.98
<sup>241</sup> Am	8.56 10 <sup>-1*</sup>	8.715 10 <sup>-1*</sup>	1.02
<sup>79</sup> Se	4.55 10 <sup>-5*</sup>	2.437 10 <sup>-4*</sup>	5.36
<sup>90</sup> Sr	4.59 10 <sup>1*</sup>	4.754 10 <sup>1*</sup>	1.04
<sup>99</sup> Tc	9.59 10 <sup>-3*</sup>	1.052 10 <sup>-2*</sup>	1.10
<sup>126</sup> Sn	1.25 10 <sup>-4*</sup>	5.579 10 <sup>-4*</sup>	4.46
<sup>135</sup> Cs	4.16 10 <sup>-4*</sup>	4.395 10 <sup>-4*</sup>	1.06
<sup>137</sup> Cs	6.71 10 <sup>1*</sup>	6.702 10 <sup>1*</sup>	1.00

<sup>a</sup>Concentrations in mg/g UO<sub>2</sub>.

\*, Denotes activity values; expression in mCi g<sup>-1</sup> UO<sub>2</sub>.

centration there is a clear improvement in the results.

From Table 4 it is also apparent that the calculated and measured activities of <sup>79</sup>Se and <sup>126</sup>Sn widely differ. This turns out to be the case for all calculations. Therefore, this cannot be caused by the update of cross sections, yield or decay data. Further investigations in the framework of the benchmark test will be needed to reveal the cause of this difference.

In these ORIGEN-S runs the cross sections and the spectral parameters are constant over the whole life cycle of the fuel. As the neutron spectrum will change continuously due to the changing composition of the fuel, the three-group cross sections and the spectral parameters used in the ORIGEN-S calculation should be regularly recalculated in a cell calculation. To this end the SAS2H module is available in the SCALE system. However, in The Netherlands an adapted version of this depletion module is developed, named SAS6 (De Leege et al., 1995), in which the cell calculation by the XSDRNPM-S neutron transport code is replaced by the much faster collision probability code WIMS-D. To see the effect of a

regular update of neutron cross sections a SAS6 run was performed using a 28 energy group library, which was condensed from a 172 fine group cross-section library based on JEF2.2 data. During each irradiation cycle the three group cross sections of selected nuclides taken into account explicitly in the cell calculation (<sup>234</sup>U, <sup>235</sup>U, <sup>236</sup>U, <sup>238</sup>U, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu, <sup>242</sup>Pu, <sup>135</sup>Xe and <sup>149</sup>Sm) are recalculated from a condensation with the calculated neutron spectrum. For all other nuclides in the burn-up calculation the cross sections are taken from the generally updated library. However, as the spectral parameters are also recalculated at every cell calculation, the one-group cross sections actually used in ORIGEN-S will also change for all nuclides.

Table 5 shows the results of the SAS6 run. Except for <sup>79</sup>Se and <sup>126</sup>Sn the results clearly improved and are all close to the experimental value.

## 10. Summary and conclusions

Data libraries for the ORIGEN-S fuel depletion code have been updated with cross sections based

on the JEF2.2 evaluated nuclear data file and the EAF3 neutron activation file. For both a LWR and a LMFBR cross sections of 517 light elements, 65 actinides and 319 fission products have been renewed or added to the libraries. Multi-group cross sections have been condensed to three-group cross sections using the neutron spectrum of a typical PWR for fuel halfway its final burn-up. For a typical LMFBR one-group cross sections have been derived, also halfway its final burn-up.

The effects of new cross sections have been evaluated by comparing the nuclide densities of actinides and fission products after four irradiation cycles in a PWR up to a burn-up of 27.35 MWd kg<sup>-1</sup> U and after another 5 years cooling time, according to the specifications of a burn-up credit criticality benchmark (Brady, 1993).

The effect of using the updated cross sections amounts up to 40% for the higher transuranics and is also considerable for specific fission products like <sup>86</sup>Rb, <sup>134</sup>Cs, <sup>136</sup>Cs and several Sm-isotopes.

The radioactive-decay data of all nuclides, including the half life, the probabilities for different decay types and the recoverable energy in decay, are updated using the JEF2.2 special purpose file with radioactive-decay data. The effects of this update are in general small, except for some Te-isotopes and many minor fission products. For <sup>241</sup>Am and <sup>242</sup>Cm they amount to 2%.

For those fissionable nuclides allowed in the ORIGEN-S fission-product library the yields of the fission products from the JEF2.2 special purpose fission-product yield data file are included. This resulted in the addition of 201 fission products to the library. The effect of updated yields generally amounts to several per cent with higher values for specific fission products like <sup>86</sup>Rb, <sup>109</sup>Ag, <sup>126</sup>Sn, <sup>136</sup>Cs, <sup>155</sup>Gd and some isomeric states of Te.

Also the fission energy and capture energy values contained in the source code of ORIGEN-S were updated from JEF2.2 data. This leads to a 0.4–0.5% increase in neutron flux at the same power and a likewise increase in fission-product concentration. At the same time the <sup>235</sup>U concentration decreases with this amount while the higher actinides show an increase up to 2%.

Finally, it was also investigated what the effect is of neglecting the fission-product yield of other fissionable nuclides than those for which yield data are provided in the ORIGEN-S library. To this end the ORIGEN-S source code was changed to substitute the <sup>239</sup>Pu yields for those nuclides. This generally gives an increase in fission-product concentrations of 0.1–0.4%. As this increases enormously the number of non-zero elements in the transition matrix, this change in the source of the ORIGEN (and COUPLE) code is not recommended.

As a general conclusion one can state that updating nuclear data for depletion calculations considerably affects the concentrations of the actinides and specific fission products at the end of fuel life time. This is especially due to the cross-section update. Updating the capture and fission reaction energies gives a small but systematic decrease in <sup>235</sup>U concentration and a systematic increase of fission-product and higher actinide concentrations.

Calculation of nuclide concentrations over the whole life time of UO<sub>2</sub> fuel with constant cross sections gives satisfactory results for most nuclides compared with measurements.

If the spectral parameters THERM, RES and FAST and the cross sections for the major U- and Pu-isotopes are recalculated regularly during an irradiation cycle in a pin-cell criticality calculation, very good correspondence with measured values for nuclide concentrations or activities are obtained. This validates the updated ORIGEN-S libraries.

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