Intelligent nuclide selection capability in the reactor physics and inventory calculation code SERPENT 2

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Abstract

SERPENT is a user-friendly reactor physics and inventory calculation code developed at VTT and used by many organisations around the world [1]. Since the neutron transport of SERPENT is based on the continuous-energy Monte Carlo method, it is well capable of calculating the nuclide inventories for all kinds of reactors and fuels – from ordinary LWRs to fast reactors – with high accuracy.

The output of SERPENT 2 is versatile, containing for example total and nuclide-wise masses, activities, radiotoxicities and heat productions in the system at each time step. However, the results are only provided for nuclides specifically chosen by the user. For an experienced reactor physicist the nuclides of interest in a problem may be obvious, but a user with less experience might face serious problems when trying to identify the important nuclides in the vast selection.

To further increase the usability of SERPENT, a new intelligent nuclide selection capability for the output of SERPENT 2 has been developed and implemented in the code. The new capability has been designed with especially final disposal, transmutation and other fuel cycle-related analyses kept in mind. The new routine selects the output nuclides on the basis of their contributions to a user-chosen property. For example, the user may request results for the 10 nuclides with the highest decay heat production. The new version of SERPENT also includes a selection of pre-defined nuclide groups for different purposes involving, for instance, the nuclides with a high-migration probability and the minor actinides. Also, the nuclide list used in COSI6 is included to provide for easy importing of materials from SERPENT to COSI6.

This paper describes the implementation of the new nuclide selection capability in SERPENT 2. The applicability of SERPENT in inventory calculations is also discussed through practical examples.

Introduction

Resolution of the time evolution of nuclide inventories in the nuclear fuel is an essential part of each fuel cycle analysis. During fuel irradiation in a reactor its composition changes rapidly, which strongly affects the performance of the reactor on a few-year time scale. After removal from the reactor, the fuel is cooled down in interim storage pools for several years and is then either reprocessed or put into final disposal. At all times, from the very first exposure to neutron flux to tens of thousands of years in final disposal, the constantly changing nuclide composition of the fuel material ultimately defines the behaviour of the fuel, including for example its radiotoxicity and decay heat production properties. Of course, the nuclide compositions themselves are also of great interest in fuel cycle analyses when closed nuclear fuel cycles are considered.

There are a few different approaches to solving the time evolution of the nuclear fuel and the reactor. Some codes, like CESAR and ORIGEN (when used standalone), rely on pre-generated, representative transmutation cross-sections for different kinds of nuclear systems. With these codes it is very fast and easy to calculate the nuclide compositions in various situations. The accuracy of the results depends on how well the pre-generated cross-sections correspond to the modelled nuclear system [1] [2].

In the case of exotic reactor designs or if a very high accuracy is required, the solution of neutronics must be introduced in the calculation procedure to provide accurate, time-dependent transmutation cross-sections. The neutronics solution can be made using either deterministic methods or the Monte Carlo method. In general, the advantage in deterministic methods is a faster calculation time, while the calculation intensive Monte Carlo method is a very general method giving good results for all kinds of nuclear systems regardless of their geometry, neutron spectrum or, for instance, temperature.

Inventory calculation codes EVOLCODE2 and MonteBurns use the Monte Carlo transport code MCNP to provide the spectrum-weighted transmutation cross-sections and the inventory calculation code ORIGEN to calculate the changes in the fuel composition. This combination provides for versatile and accurate modelling of the time evolution of a wide variety of nuclear systems [3-5]. Similar inventory calculation functionality can be found within the SERPENT code, a Monte Carlo reactor physics and burn-up calculation code, which is experiencing growing success. For those already familiar with SERPENT, it is the natural tool to be used in fuel cycle analyses. The progressive burn-up methodology of SERPENT can also be used to provide results for comparison with other codes [6] [7].

It was noticed that the usability of SERPENT in fuel cycle related analyses could be easily increased by introducing a few new and convenient features for selecting the nuclides of interest in the SERPENT output. The new features are of great help for SERPENT users and significantly ease the usage of SERPENT in conjunction with the fuel cycle scenario code COSI6 [8].

Serpent as an inventory calculator

SERPENT is a Monte Carlo reactor physics code that has been developed at VTT Technical Research Centre of Finland since 2004 by Jaakko Leppänen. The source code for SERPENT 1 can be obtained free of charge from the OECD/NEA Databank or the RSICC for non-commercial research and educational use. Currently, the user community of SERPENT 1 spans more than 170 users in 72 universities and research organisations around the world. SERPENT 2, which is the code version used in this work, is a new development version of the SERPENT code and is at the moment only available to a limited group of licensed beta-testers [6].

It should be emphasised that SERPENT is, above all, a reactor physics code instead of a full-fledged inventory calculator. The code, however, contains a burn-up calculation capability based on the CRAM method, which makes it possible to calculate the time development of the nuclide inventory during fuel irradiation, storage and final disposal [9]. In a burn-up calculation the neutron transport calculation first produces the one-group transmutation cross-sections in the system. These cross-sections are then used by the burn-up calculation module to solve the Bateman equations, resulting in the nuclide compositions at the beginning of the next burn-up step [10]. The procedure is repeated until the whole irradiation and/or decay history is covered.

The neutron transport of SERPENT is based on the continuous-energy Monte Carlo method. Consequently, it is well-suited for the modelling of all kinds of reactors and fuels, practically without any approximations. The price of the generality is a prolonged calculation time compared to a conventional inventory calculation with pre-generated cross-sections or a calculation based on deterministically resolved transmutation cross-sections: a typical bundle-level burn-up calculation with SERPENT takes several hours of CPU time (or about 20 minutes per burn-up step), almost all of which is spent in the solution of neutronics. Fortunately, the calculation effort can, nowadays, be efficiently divided among multiple processors. Due to several optimisation tweaks utilised in SERPENT, the calculation times are also very competitive compared to many other codes or code couplings with the same basic methodology.

SERPENT uses ACE-formatted cross-section data while fission yield and radioactive decay data are read from ENDF-formatted files. Since the ACE-formatted cross-section libraries can be rather straightforwardly processed from the ENDF nuclear data files using NJOY, the nuclear data libraries of SERPENT are easy to change for comparisons with older evaluations or to update as new evaluations are published [11]. SERPENT utilises all of the available decay, fission yield and cross-section data, which in the case of JEFF-3.1.1 nuclear data files corresponds to about 300 nuclides in the transport and altogether 1 550 nuclides in the burn-up calculation. The number of nuclides involved in the calculation is not as high as with some inventory calculation codes like ORIGEN, but the number is still considered sufficient for most practical purposes [2]. The number of available nuclides also keeps increasing along with the completion of the nuclear data libraries.

In summary, SERPENT can be used to calculate inventories of all kinds of nuclear systems with only the system description together with basic nuclear data in ENDF and ACE formats as input.

New intelligent inventory nuclide selection capabilities

Both SERPENT 1 and SERPENT 2 are well-capable of making inventory calculations. The standard output includes total and material-wise atomic densities, masses, spontaneous fission rates, decay heat productions and radiotoxicities, which is sufficient for most analyses. For each of these quantities, contributions of so-called inventory nuclides are printed out separately in addition to the total value, which is simply the sum over all nuclides. Obviously, the nuclide-wise contributions play a major role when making fuel-cycle analyses – especially when fuel reprocessing is involved.

In the current versions of SERPENT 1 and 2, the inventory nuclides are set manually in the SERPENT input, one by one, by the user. This is a little bit troublesome, because in many cases the user does not know which the nuclides of interest really are or the list might be both long and problem dependent. The issues are emphasised for inexperienced reactor physicists.

To increase the user-friendliness of SERPENT, the nuclide inventory selection capabilities of SERPENT are developed further in this work. In the following, two new ways of including nuclides in the inventory are introduced, namely hard-coded nuclide

lists and a capability of automatically including top-*N* contributors of a quantity to the inventory.

Hard-coded lists of important nuclides

The main reason for implementing the hard-coded nuclide lists into SERPENT 2 was to ease the importing of materials from SERPENT to COSI6. However, the property can also be taken advantage of by introducing predefined lists of nuclides that are found important in different kinds of analyses. The nuclide lists may spare a lot of manual work in the creation of SERPENT input files.

The inventory nuclides are chosen in the SERPENT input by command "set inventory" followed by the nuclide ZAI identifiers. For example, command

```
set inventory
922350
922380
```

includes the two most common uranium isotopes in the inventory. With the new property implemented, the identifiers of individual nuclides can be replaced by keywords that correspond to a list of nuclides. For example,

```
set inventory
lanthanides
minoractinides
```

includes all lanthanides and minor actinides found in JEFF-3.1.1 data libraries in the inventory. A complete list of currently implemented keywords with explanations and references for the corresponding nuclides are listed in Table 1.

Table 1: Keywords and their explanations for hard-coded nuclide lists in SERPENT 2

Keyword	Explanation	Reference
accident	20 nuclides with significant health impact & high migration probability in accident conditions.	[12]
actinides	All actinides for which neutron cross sections are available in the JEFF-3.1.1 nuclear data libraries. Altogether 63 nuclides.	[13]
Burn-up credit	31 nuclides that are commonly considered in burnup credit criticality safety analyses.	[14]
COSI6	Nuclide list used by fuel cycle scenario code COSI6.	[8]
lanthanides	All lanthanides for which neutron cross sections are available in the JEFF-3.1.1 nuclear data libraries. Altogether 62 nuclides.	[13]
Long-term	111 relevant radionuclides in long-term waste analyses.	[15]
Minor actinides	All minor actinides for which neutron cross sections are available in the JEFF-3.1.1 nuclear data libraries. Altogether 39 nuclides.	[13]

Additionally, to provide for automatic importing of fuel compositions from SERPENT to COSI6 (the final imposed content –option), a BASH script for converting Matlab-formatted SERPENT output into COSI6-compatible xml format was written.

Top-N contributors

Often the user of an inventory calculator code is interested in the most important contributor nuclides to some quantity, for example decay heat production. An experienced reactor physicist might know the nuclides of interest in each situation

beforehand, but to facilitate analyses by less-experienced users and to avoid human errors, an automatic inventory nuclide selection property was implemented in SERPENT 2.

The nuclide selection property ensures that top-*N* contributors of a quantity, chosen by the user, are included in the nuclide inventory at each burn-up step. The traceable quantities involve mass, activity, spontaneous fission (keyword sf), decay heat (decheat) together with inhalation and ingestion radiotoxicities (ingtox, inhtox). In practice, the top-*N* property can be activated by using the keyword "top" in the inventory definition followed by the number *N* and the quantity keyword. For example, an inventory definition

```
set inventory
922380
lanthanides
top 5 sf
top 5 decheat
```

would include the 5 most important spontaneous fission source nuclides and the 5 most important decay heat producers in the inventory, in addition to ^{238}U and the lanthanides.

Demonstration of the new properties

The new property is demonstrated by examining the decay heat production in two different, simple but realistic nuclear systems: a Gd-doped LWR fuel assembly and a SFR bundle. Both of the systems are first burned to a total burn-up of 60 MWd/kgHMi using 44 burn-up steps and then cooled down for 100 000 years using 47 decay steps. The burn-up equations are solved using the CRAM method and a first order predictor-corrector method is also utilised in the time integration. Three million active neutron histories were calculated in each transport cycle.

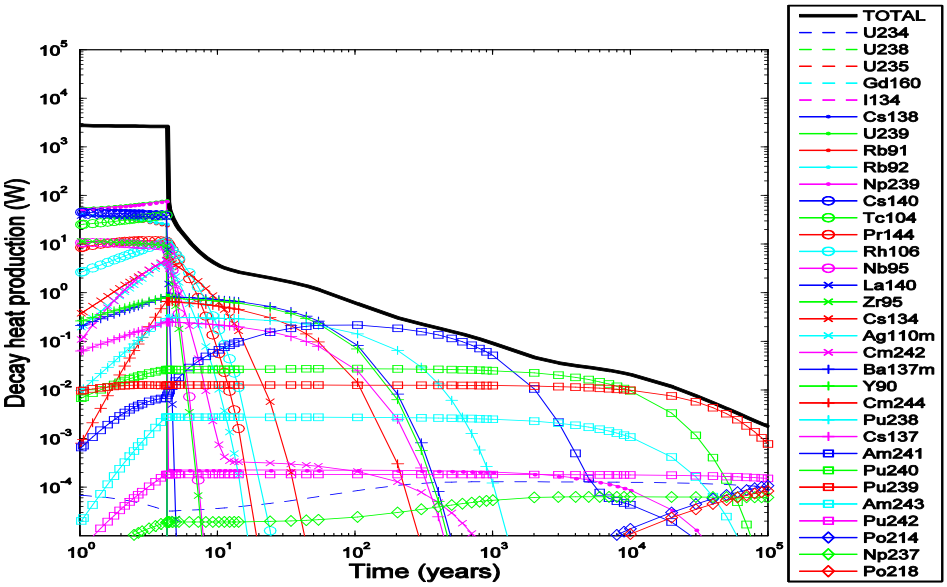
The calculations were performed using a computer with 12 3.47 GHz Intel Xeon cores and 48 Gb of memory. The calculation time was 3 h 20 min in the LWR case and 2 h 20 min in the SFR calculation.

LWR assembly

The LWR case involves an ordinary 17x17 pressurised water reactor (PWR) bundle with 16 of the rods doped with Gd. The bundle is based on a NEA benchmark [16]. SERPENT input for the problem can be found in the SERPENT website [1]. For the present calculation the input was, however, slightly modified by disabling the energy-grid thinning and removing the cut-offs for burn-up calculation. An average power density of 38.6 W/g was used in the power normalisation, corresponding to a total power of 45 870 W/cm.

The five most important decay heat producers are included in the inventory by using the new top-*N* property described above. The total and nuclide-wise decay heat productions for the whole calculation period are plotted in Figure 1. Since the calculation is performed for an axially infinite fuel bundle, the results are expressed in watts per axial centimetre (of the bundle). The left-hand side of the plot represents the irradiation period and after about 3.5 years of operation the fuel is removed from the reactor. After removal, the fuel is no longer exposed to neutron flux and, hence, only decay reactions take place.

Figure 1: Total decay heat production and the nuclide-wise productions of the top-5 decay heat producers in the PWR case



The decay heat production is expressed per one axial cm of the fuel bundle. This plot demonstrates the usage of the new top-*N* inventory nuclide selection property in SERPENT 2.

First, it should be noted that the top-5 decay heat producers *at every burn-up step* are included in the inventory, also at the very beginning of the burn-up calculation when the fuel is still fresh. Therefore, for example, uranium isotopes 235 and 238 are shown in the list of nuclides even though their contribution to total decay heat production is negligible at relevant burn-ups. Because of the scale used, their contribution cannot be seen in the plot, either.

In the plot it can be seen that the top-5 decay heat producer nuclides are each responsible of only a few per cent of the total decay heat production when fuel is in the reactor. Hence, it is not very fruitful to track individual decay heat producer isotopes during fuel irradiation. As the reactor is shut down, the short-lived nuclides disappear suddenly and the situation becomes quite different.

About 20-40 years after removal from reactor, a time interval very relevant to final disposal analyses, the most important decay heat producers are fission products ^{137m}Ba, ^{90}Y and ^{137}Cs together with actinides ^{238}Pu and ^{244}Cm. At this point the top-5 nuclides are, together, responsible for 46% of the total heat production. After about 60 years, ^{241}Am, a decay product of ^{241}Pu, becomes the most important decay heat producer and remains the most important contributor until about 1 000 years have elapsed. Thereafter, the most important decay heat producers are plutonium isotopes 239 and 240.

SFR assembly

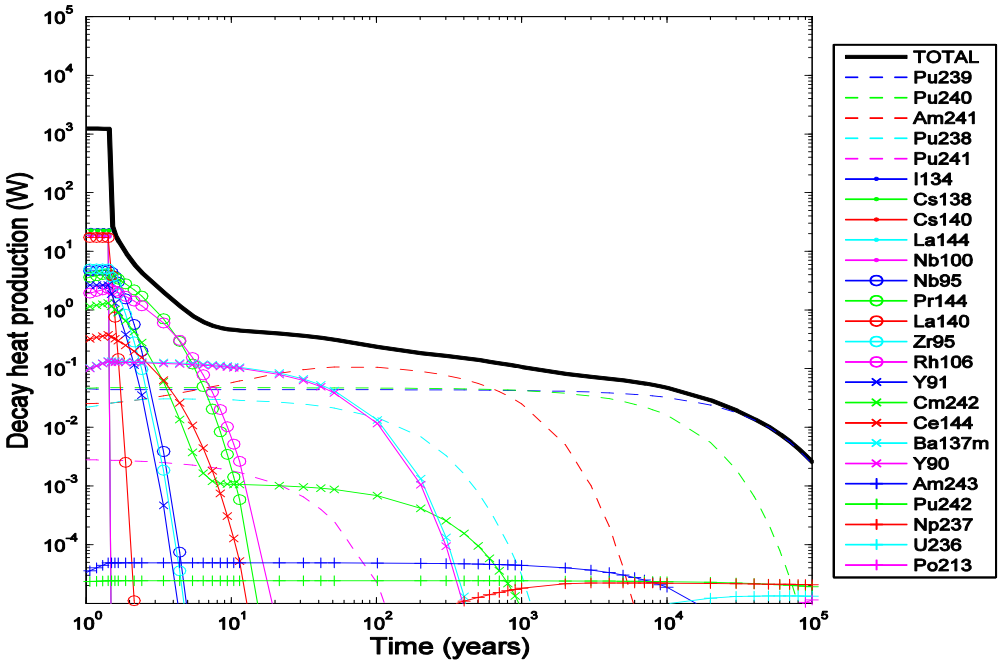
The sodium-cooled fast reactor (SFR) assembly example was taken from a compilation of inputs regularly used for the validation of SERPENT against MCNP [17]. The assembly consists of 91 fuel pins in a hexagonal lattice. The fuel material is fresh MOX fuel the isotopic composition of which is described in Table 2. The power is normalised to an average linear power of 240 W/cm for each fuel rod corresponding to a total power of 21 840 W/cm. Although the usage of the probability table treatment for unresolved region cross-sections might be reasonable for this fast spectrum system, the probability table

sampling was not used in the neutron transport calculation. Again, top-5 decay heat producers were tracked throughout the calculation. The results are plotted in Figure 2.

Table 2: Nuclide composition of the MOX fuel material in the SFR example case

Nuclide	Percentage (a-%)
^{235}U	6.374
^{238}U	21.080
^{238}Pu	0.006
^{239}Pu	4.496
^{240}Pu	1.121
^{241}Pu	0.154
^{242}Pu	0.032
^{241}Am	0.037
^{16}O	66.701

Figure 2: Total decay heat production and the nuclide-wise productions of the top-5 decay heat producers in the SFR case



By comparing this plot to Figure 1, it can be seen that the total decay heat production curve is a different shape than in the PWR case.

During fuel irradiation the proportion of the decay heat power to the total heating power is about the same in the SFR case as it is in the LWR case (5.9% in SFR case vs. 6.0% in PWR). After removal from reactor the total decay production curve differs slightly from that of the PWR case. Namely, it seems that the decay heat production drops much faster at the beginning of the cooling-down period of the SFR fuel. On the other hand, after this fast cooling period the decay heat production descends slower than in the PWR case.

In the plot it can be observed that some important decay heat producers at 20-40 years after irradiation, for example ^{137m}Ba and ^{90}Y, have significantly smaller concentrations in the SFR case. The lack of these two nuclides, together with other nuclides with similar half-lives, is the main reason for the faster drop in decay heat production.

When about 100 years have passed since the fuel irradiation, the long-lived plutonium isotopes 239 and 240 start to dominate the heat production and the domination continues until the end of the calculation period. Since the content of these isotopes is higher in SFR fuels than in PWR fuels, the decay heat production descends very slowly and, as a consequence, the decay heat production at the end of the 100 000 year calculation period is even larger than that in the PWR case, despite the lower total heat production of the SFR bundle. However, the absolute value of the decay heat production 2.5 mW/cm at the end of the calculation period is, in practice, insignificant.

Conclusions and discussion

Two new options for selecting inventory nuclides are implemented in the Monte Carlo reactor physics and the inventory calculation code SERPENT 2. The two options, namely hard-coded nuclide lists and top-*N* contributor -property, increase the usability of SERPENT 2 in fuel cycle -related analyses. As the nuclide list used by the fuel cycle scenario code COSI6 is one of the new hard-coded nuclide lists, material compositions calculated with the new version of SERPENT 2 can be automatically imported into COSI6 from the newest version of SERPENT 2.

The top-*N* contributor property makes it possible to easily recognise the most important nuclides in a problem by including the most important contributors of total mass, activity, spontaneous fission source, decay heat production or radiotoxicity in the list of inventory nuclides. Since, for example, migration probabilities of nuclides in different conditions are not included in the list of traceable quantities, it is still necessary and practical to use hard-coded nuclide lists in certain analyses.

Acknowledgements

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