The TOWER Undergraduate Research Journal
Georgia Institute of Technology
gttower.org

Development of an Algorithm for Determining Radiotoxicities of Spent Nuclear Fuel over Time

ROBERT ADAMS

Knowledge of how the radiotoxicity of spent nuclear fuel changes over time is essential when designing and evaluating spent fuel storage and treatment methods. The common method of performing calculations to obtain these data is to use standardized transmutation codes, which are computer programs with built-in solution methods to a given set of problem types. Currently widespread transmutation codes require a time-intensive process in order to prepare calculations of this nature and obtain the results in a convenient format. Furthermore, access to these transmutation codes is not universal. The purpose of this work was to use one of these codes, EASY2003, to develop a simpler and faster algorithm for solution of these time-dependent radiotoxicity changes. The basic methodology employed was to perform EASY2003 calculations separately on all relevant isotopes, and then to generate an algorithm that can amalgamate these individual results into a solution of any given fuel mixture and amount. For verification purposes, the algorithm generated was applied to a sample spent fuel mixture, and the solution was compared with a more traditional solution method generated by the transmutation code ORIGEN-ARP.

INTRODUCTION

The Global Nuclear Energy Partnership (GNEP), announced in February 2006 (Stevens, 2006), is a program lead by the United States Department of Energy intended to, among other things, incorporate fuel reprocessing in the American nuclear fuel cycle. One motivation behind this policy is to extract more energy from a given amount of mined uranium while, at the same time, reducing the amount of high level radioactive waste that must be dealt with at the end of the fuel cycle. Some important factors in determining what methods are most effective to this end are the total activity, dose rates, and heat production of given spent fuel compositions as they change over long periods of time (Stacey, 2007). Typically, standardized transmutation codes are used to obtain these data. The codes are essentially computer programs with built-in solution methods to a given set of problem types. Currently available transmutation codes, such as EASY2003 (Forrest, 2002) and ORIGEN-ARP (Bowman, S. M., Gauld, I. C, & Horwedel, J. E., 2006) can perform these calculations, but various factors can make these methods unappealing. The purpose of this work was to develop a means to more rapidly determine how the radiotoxicity of a given spent nuclear fuel composition changes over very long periods of time, up to 100,000 years in this case, and to make this method more accessible.

EASY2003 and ORIGEN-ARP are codes capable of performing calculations that are immensely more complicated and demanding than those addressed in this paper. The scenarios addressed in this paper, however, do not require the complex nature of these transmutation codes, as the case of spent nuclear fuel decaying over time does not require consideration of neutron irradiation, a factor that makes calculations of this nature far more complex and numerically intensive. If one wishes only to perform this relatively simplified case, though, some of the same challenges of setting up the codes to solve those more complex problems must be faced if EASY2003 is utilized. Firstly, access to these codes is not universal. They are maintained and distributed by centralized organizations, and access is limited by issues such as cost and export controls. These codes incorporate large amounts of isotopic cross-section data, which corre-

Contact email: gtg548x@mail.gatech.edu
Advisor: W. F. G. van Rooijen, PhD
School: Nuclear and Radiological Engineering Program
George W. Woodruff School of Mechanical Engineering
Georgia Institute of Technology

spond to the likelihood of interaction between a given neutron as it moves through a material of a given isotope. Codes such as these also tend to come with a steep learning curve that a user may not wish to overcome if he only wishes to use a given code for its more simple capabilities. Thus, the goal of this research endeavor was essentially to develop a simple and accessible solution method to this simplified case that allows one to circumvent these demanding and complicated solution methods.

The nuclear engineering software package EASY2003 was used to develop values of contribution by all relevant actinides, as the long-term characteristics of spent nuclear fuel are primarily governed by these actinides (Stacey, 2007). Actinides consist of the fifteen chemical elements between actinium and lawrencium on the periodic table of elements. Several actinides are considered relevant in this paper, because they have a propensity to build up during operation of a nuclear reactor. The contributions of these individual actinides are not interdependent, and can thus be calculated separately and later incorporated in a simple algorithm to sum these individual components based on an initial spent fuel composition (Stacey, 2007). This system characteristic, the lack of interdependence, is the basis for simplifying the process of determining spent fuel characteristics as they evolve over time; given an initial spent fuel composition, the time-dependent properties can be instantly obtained once these individual source terms are known. In order to verify the final algorithm developed by this process, the decay characteristics of a typical light-water reactor spent fuel composition were solved and compared with a solution generated by the software package ORIGEN-ARP.

Both ORIGEN-ARP and EASY2003 are capable of performing a wide array of decay and transmutation calculations, including the ones described in this paper, but they were developed independently of one-another, and thus can be seen as offering a higher degree of confidence in the event of both yielding the same result for a given calculation.

METHODS

The core function of the module FISPACT within EASY2003 is to solve the governing body of differential equations that describes the composition of a given mixture of radioactive isotopes as it changes with time. This calculation can be performed with or without the presence of an incident neutron flux. Each individual isotope present in the material mixture is represented by a single differential equation of the form in Equation 1 (Forrest, 2002):

$$\frac{dN_i}{dt} = -N_i(\lambda_i + \sigma_i\phi) + \sum_{j\neq 1} N_j(\lambda_{ij} + \sigma_{ij}\phi) + S_i \qquad (1)$$

$$S_i = \sum_k N_k \sigma_k^f \phi Y_{ik}$$

Where:

N_i is the amount of isotope *i* at time *t* (cm^{-3}).
λ_i is the decay constant of isotope *i* (s^{-1}).
λ_{ij} is the decay constant of isotope *j* producing *i* (s^{-1}).
σ_i is the total cross section for reactions in *i* (cm^2).
σ_{ij} is the reaction cross section for reactions on *j* producing *i* (cm^2).
σ_k^f is the reaction cross section (f indicating fission) for reactions on actinide *k* (cm^2).
ϕ is the neutron flux (n cm^{-2} s^{-1}).
S_i is the source of isotope *i* from fission.
Y_{ik} is the yield of isotope *i* from the fission of isotope *k*.

In the case of spent nuclear fuel decay, as is being addressed here, the neutron flux is zero, since there is no source of neutrons as in an online nuclear reactor, and the Equation 1 can be simplified to Equation 2:

$$\frac{dN_i}{dt} = -N_i\lambda_i + \sum_{j\neq 1} N_j\lambda_{ij} \qquad (2)$$

The important aspect of the EASY2003 solution method that allows it to be used in the generation of a simpler, faster algorithm for solving spent fuel decay characteristics is that the solution to each isotope's differential equation is not dependent upon the solution of any other isotope. Once the solution for each isotope *j* is known for a given amount of elapsed time *i*, the total activity, or number of decays per second, of a given spent fuel mixture can be calculated as in Equation 3:

$$A_{total,i} = M\sum_j f_j A_{ij} \qquad (3)$$

Where :

$A_{total,i}$ is the total activity of the fuel composition at time *i* (Bq).
M is the total mass of the fuel (kg).
f_i is the mass fraction of isotope *j* (unitless).
A_{ij} is the activity of isotope *j* at time *i* (Bq/kg).

The same solution method can be applied to the values of heat output and gamma ray dose rate at a given point in time. If all relevant values are already calculated, the decay characteristics (total activity, heat generation, gamma dose rate) of any fuel composition can be very simply obtained by Equation 3 without the need to re-solve the entire set of differential equations described in Equation 1.

RESULTS

Solutions were obtained at one-hundred steps of elapsed time, logarithmically distributed from zero to one-hundred thousand years. Results were obtained for all actinides with long enough half-lives such that they are relevant to long-term fuel storage,

as listed in Table 1. A half-life longer than approximately two years is considered relevant, as spent-fuel is allowed to lie dormant for much longer than this time period before being considered for long-term waste storage.

Sample total activity solutions can be seen for the individual isotopes of Am-241, Cm-244, and Np-237, summed with their respective progeny, in Figure 1. These are three of the most important isotopes in spent nuclear fuel (Stacey, 2007). The gamma dose rate data for these isotopes can be seen in Figure 2 and the heat output data can be seen in Figure 3.

For purposes of verification, the overall decay characteristics of a typical light water reactor spent fuel composition were calculated using the algorithm generated from EASY2003 and then compared with the results obtained when using ORIGEN-ARP with the same initial composition. This is intended to be a wholesale comparison between the method generated and the results obtained from EASY-2003 and an alternative transmutation code, ORIGEN-ARP. Presuming that the solution method detailed within this paper is exactly representative of the more complex EASY-2003 solution method, minor differences between it and ORIGEN-ARP results are only indicative of differences between the two codes, not a degree of error inherent in the simplified solution method. Because of this, minor differences can only be explained by delving deep into the workings and merit of EASY2003 and ORIGEN-ARP, a task far beyond the scope of this paper. This is also the reason why one composition is used as representative, as minor changes in the composition would only indicate a similar style of error, not one indicative of the simplified solution method. In general, the comparison only offers proof that a significant user error was not made in generating the algorithm, something that would only appear in the form of one or more orders of magnitude difference. The spent fuel mixture used was generated using the sample fuel burn-up calculation included with the TRITON module within the SCALE5 software package (Stevens, 2006). SCALE5 is a nuclear code package that includes a wide array of modules for modeling a plethora of nuclear engineering systems, including a case of light-water reactor fuel irradiation and neutronic behavior. The spent fuel composition used can be seen in Table 2.

The total activity and total heat output obtained from the algorithm generated from EASY2003 and the results obtained from ORIGEN-ARP can be seen in Figures 4 and 5. The re-

Am-241	Cf-249	Cm-242	Cm-246	Np-236	Pu-238	Pu-242	U-234
Am-242	Cf-250	Cm-243	Cm-247	Np-237	Pu-239	Pu-244	U-235
Am-243	Cf-251	Cm-244	Cm-248		Pu-240		U-236
	Cf-252	Cm-245	Cm-250		Pu-241		U-238

Table 1. List of relevant isotopes accounted for in final algorithm.

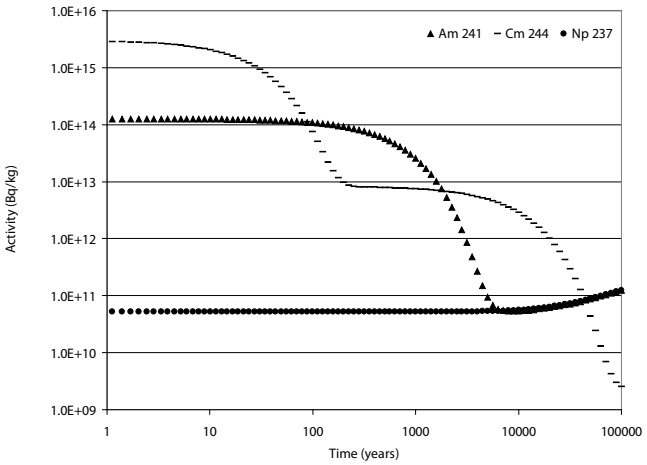

Figure 1. Activity of the sum of the given isotope and its progeny for three particularly relevant actinides, intended to be a sample of the twenty-seven actinides solved in this study.

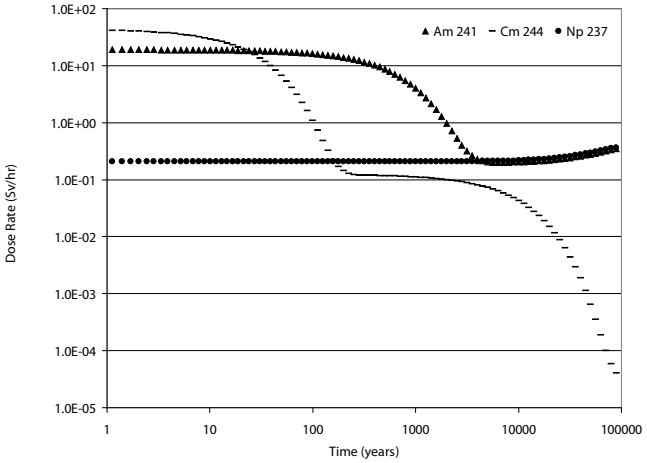

Figure 2. Gamma dose rate of the sum of the given isotope and its progeny for three particularly relevant actinides, intended to be a sample of the twenty-seven actinides solved in this study.

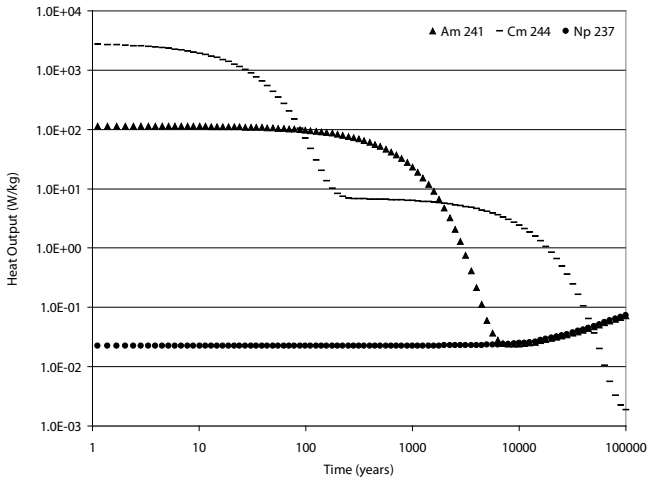

Figure 3. Heat output of the sum of the given isotope and its progeny for three particularly relevant actinides, intended to be a sample of the twenty-seven actinides solved in this study.

Isotope	Mass fraction	Isotope	Mass fraction
U-234	2.74×10^{-4}	Np-237	3.95×10^{-4}
U-235	1.85×10^{-2}	Am-241	6.18×10^{-5}
U-236	4.41×10^{-3}	Am-243	4.64×10^{-5}
U-238	9.66×10^{-1}	Cm-242	3.96×10^{-6}
Pu-238	1.02×10^{-4}	Cm-243	1.67×10^{-7}
Pu-249	6.95×10^{-3}	Cm-244	9.82×10^{-6}
Pu-240	1.87×10^{-3}	Cm-245	3.62×10^{-7}
Pu-241	1.20×10^{-3}	Cm-246	1.70×10^{-8}
Pu-242	2.61×10^{-4}	Cm-247	1.60×10^{-10}

Table 2. Example spent fuel composition.

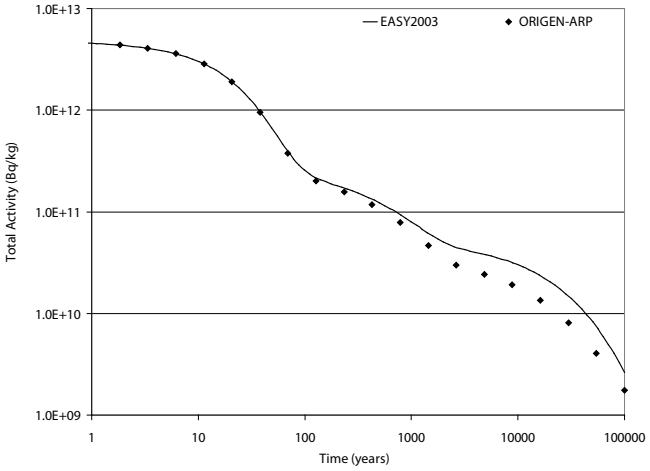

Figure 4. Comparison between the solution method employed in this study and that of ORIGEN-ARP for a representative fuel mixture in determining total activity.

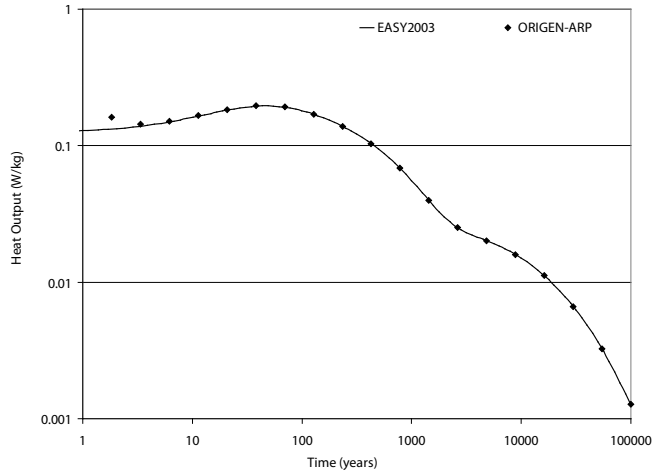

Figure 5. Comparison between the solution method employed in this study and that of ORIGEN-ARP for a representative fuel mixture in determining heat output.

sults obtained from ORIGEN-ARP are nearly identical to those obtained from the EASY2003-based algorithm, and the differences are small enough to strongly indicate that no error was made in the algorithm's development.

DISCUSSION

The algorithm for solving long-term decay characteristics of spent nuclear fuel previously outlined was proven correct by the very close correlation with a more traditional solution in the case of a typical spent-fuel composition. As results were obtained for 3 characteristics (total activity, heat generation, gamma dose rate) of 27 isotopes relevant to spent nuclear fuel at 100 points of elapsed time, the solutions of nearly any spent fuel composition can be readily obtained by the very simple summation in Equation 3. This algorithm has the potential to be very useful in future research corresponding to fuel reprocessing in accordance with the goals of GNEP. As different breeder reactor and chemical reprocessing scenarios are evaluated, the ultimate costs and benefits related to long-term fuel storage needs can be instantly and simply acquired when composition before and after a given fuel treatment is known.

Aside from offering a solution method employing benefits previously outlined to those wishing to perform calculations of this nature, this simpler and more easily accessible form may encourage researchers to perform these calculations at times when they otherwise would not. These may include times when spent fuel radiotoxicity is not the primary issue of concern for a given research endeavor, but adding such calculations would be beneficial. The final form of the results outlined in this project was an exhaustive Microsoft Excel 2007 spreadsheet. This is a format easily understood and employed by most researchers, though the data could be transferred to a variety of other forms, depending on the needs of those wishing to use them. It is clear that entering a composition into a Microsoft Excel 2007 spreadsheet is a simpler and more efficient method for acquiring and becoming proficiencient in the use of an esoteric transmutation code such as EASY2003 or ORIGEN-ARP.

ACKNOWLEDGMENTS

The author would like to thank Dr. Cassiano de Oliveira for introducing him to the world of nuclear engineering research, Dr. Wilfred van Rooijen for his benevolent guidance and counsel on this and other projects, and his parents for everything else.

REFERENCES

Bowman, S. M., Gauld, I. C., & Horwedel, J. E. (2006). *ORIGEN-ARP: Automatic Rapid Processing for Spent Fuel Depletion, Decay, and Source Term Analysis*. ORNL/TM-2005/39. Revision 5.1. Vol. I, Book 2, Sect D1.

DeHart, M. D. (2006). *TRITON: A Two-Dimensional Transport and Depletion Module for Characterization of Spent Nuclear Fuel*. ORNL/TM-2005/39. Revision 5.1. Vol. I, Book 3, Sect T1.

Forrest, R. A. (2002). *FISPACT-2003: User Manual*. EASY Documentation Series. UKAEA FUS 485

Stacey, W. M. (2007). Fuel Burnup. In *Nuclear Reactor Physics* (pp. 197-248). WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

Stevens, C. (2006). *Department of Energy Announces New Nuclear Initiative*. Retreived January 10, 2008, from: http://www.energy.gov/news/3161.htm