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Computing Atomic Density Changes of Material Composition in Operation of the Nuclear Reactor Core

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Abstract

The present work investigates an appropriate way to calculate the 1700 atomic density changes in the reactor operations. To automate this procedure, a computer program has been designed by C#. This program suggests a way to solve this problem which is based on the solution system of differential equations (Bitman) that it is designed according to Runge-Kutta Fehlberg method. The designed software is based on the high ability to calculate the material depletion with constant flux and constant power condition. The software inputs included, reactor power, computation time, initial and final time, determine of Taylor series order in calculation time depended flux, determination of time unite, specifyingmaterial composition of the reactor core at initial condition consist of light radioactive material, heavy and fission products, determining the order in the accuracy of calculations, applying the decay constants library, cross section database, the amount of generated thermal energy by various material decay ,determining the type of calculations at point of view constant flux or constant power. Finally, the atomic density of light, heavy materials and fission products at various times of reactor operation were calculated with high accuracy as the out puts of this program. At last, it is worth to say that we proposed a new approach for the use the Runge-Kutta Fehlberg method to compute atomic density changes of material composition of the reactor core which lead us to achievement a high ability tool to solve the above problem.

Keywords: *Atomic density, Runge-Kutta Fehlberg method, Reactor core, Burn up.*

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Introduction

The long-term changes in the properties of a nuclear reactor over its lifetime are determined by the changes in composition due to fuel burn-up and the manner in which these are compensated. [1]It is extremely important to monitor the isotopic composition in the core during reactor operation, since changes in core composition can affect core multiplication as well as flux and power distributions. Certainly, the prediction of fuel depletion and conversion in the core is essential for the determination of fuel loading requirements. Furthermore certain of the fission product nuclei, or their progeny following radioactive decay, are characterized by extremely large absorption cross sections and hence may significantly affect the reactivity of the core. [1, 2, 3]

The initial composition of a fuel element will depend on the source of the fuel. for reactor operating on the uranium cycle , fuel developed directly from natural uranium will contain a mixture of 234U ,235U and 238U , with the fissile 235U content varying from 0.72 % (from natural uranium) to more than 90% , depending on the enrichment. Recycle fuel from reprocessing plants will also contain the various isotopes produced in the transmutation-decay process of uranium. [1, 3] During the operation of a nuclear reactor a number of changes occur in the composition of the fuel. The various fuel nuclei are transmuted by neutron capture and subsequent decay. The fission event destroys a fissile nucleus. Of course and in the process produces two inter mediate mass fission products.

The most commonly used measure of fuel burn up is the fission energy release per unite mass of fuel. The fission energy release in megawatt-day divided by the total mass (in unite of 1000 kg or 1 tone) of fuel nuclei in the initial loading is referred to as megawatt-days per tone. [1, 2, 4]

Concentrations of the various fuelisotopes in a reactor are described by a coupled set of production-destruction equations. In determining the time dependence of nuclide concentrations, we solve the following equation: [1, 5]

$$\frac{dN_i}{dt} = \sum_{j=1}^{N} l_{ij}\lambda_j N_j + \sum_{j=1}^{N} f_{ij}\sigma_{aj} N_j \varphi - (\lambda_i + \sigma_{ai}\varphi)N_i \qquad (1)$$

Where, l_{ij} is the fraction of constant decay of the isotope j-th that are eventuated to product the isotope i-th, f_{ij} is the fraction of the absorption cross section of isotope i-th, N is the number of isotope, λ_j,λ_i are constant decay of isotope i-th and j-th, σ_{ai} is the absorption cross section of isotope i-th, N_i is atomic density of isotope i-th. Equation (1) can be integrated to determine composition changes over the life time of the reactor core loading if the dependence of the flux is known.a neutron flux distribution is calculated for the beginning of cycle composition and critical control rod position or soluble boron concentration,

and flux distribution is used to integrate the composition Eq.1 over a depletion time step. [1, 4, 6, 7, 8, 9, 10, 11]

This paper, Equation (1) is written for a homogeneous medium that contain a space-energy-averaged neutron flux, ϕ, with flux-weighted average cross sections, representing the reaction probabilities. In reality, the flux as a function of space, energy and time is depending on the nuclide concentrations. We assumed the space-energy-averaged flux can beconsidered constant over time steps, **Δt**. Similarly, we assumed a single set of flux-weighted neutron cross sections is adequate **to** use over the entire fuel exposure time. For a given time step, these assumptions are necessary if Eq.1 is to be treated as a first-order, linear differential equation. [5, 6, 12, 13]

Method of calculations

To solve the Equation.1, we used Runge-Kutta Fehlberg method**:**

$$y_{i+1} = y_i + (\frac{37}{378}k_1 + \frac{250}{621}k_3 + \frac{125}{594}k_4 + \frac{512}{1771}k_6)h \qquad (2)$$

$$y_{i+1} = y_i + (\frac{2825}{27648}k_1 + \frac{18575}{48384}k_3 + \frac{13525}{55296}k_4 + \frac{277}{14336}k_5 + \frac{1}{4}k_6)h$$

$$k_1 = f(x_i, y_i)\ k_2 = f(x_i + \frac{1}{5}h, y_i + \frac{1}{5}k_1 h)\ k_3 = f(x_i + \frac{3}{10}h, y_i + \frac{3}{40}k_1 h + \frac{9}{40}k_2 h)$$

$$k_4 = f(x_i + \frac{3}{5}h, y_i + \frac{3}{10}k_1 h - \frac{9}{10}k_2 h + \frac{6}{5}k_3 h)\ k_5 = f(x_i + h, y_i - \frac{11}{54}k_1 h + \frac{5}{2}k_2 h - \frac{70}{27}k_3 h + \frac{35}{27}k_4 h)$$

$$k_6 = f(x_i + \frac{7}{8}h, y_i + \frac{1631}{55296}k_1 h + \frac{175}{512}k_2 h + \frac{575}{13824}k_3 h - \frac{44275}{110592}k_4 h + \frac{253}{4096}k_5 h)$$

By changing the value of i = 0,1,2…in y_i value, we achieved to approximate value of y_{i+1}.

To calculate the value of the error, difference between the fourth and fifth order of relations is computed. Therefore, the error relation is demonstrated as follow:

$$Error = \left(\frac{2825}{27648} - \frac{37}{378}\right)k_1 h + (0-0)k_2 h + \left(\frac{18575}{48384} - \frac{250}{621}\right)k_3 h + \left(\frac{13525}{55296} - \frac{125}{594}\right)k_4 h + \left(\frac{277}{14336} - 0\right)k_5 h + \left(\frac{1}{4} - \frac{512}{1771}\right)k_6 h \qquad (3)$$

It should to be mention that setting of the h value is based on equation.3 and the desired accuracy for this problem. Therefore, we explain this approach in Runge-Kutta Fehlberg method for different orders as below:

If the initial value of 'a' and the final value of 'b' exists, therefore, at the first we consider a value of 'h'. Then, we calculate the error and the new value of by. These values of the error are compared together to set the value of the 'h'. In this regard, n is order of Runge-Kutta Fehlberg method and $\Delta_{present}$,$h_{present}$ are the current step and error and $_{\Delta new}$,h_{new} are the new step and error, respectively [6].

$$\left(\frac{h_{new}}{h_{present}}\right)^{n+1} = \left|\frac{\Delta_{new}}{\Delta_{present}}\right| \qquad (4)$$

In the Table 1, the error rate in the solution of ordinary differential equations consists of the Euler, Improved Euler, and Runge-Kutta Fehlberg method is shown.

Table 1. Comparison of computational methods for ordinary differential equations in terms of accuracy [13].

Method	Function Evaluation	Global Error	Local Error
Euler	1	$O(h)$	$O(h^2)$
Improved Euler	3	$O(h^2)$	$O(h^3)$
RK	4	$O(h^4)$	$O(h^5)$
RKF	6	$O(h^5)$	$O(h^6)$

It is worth to say that the Runge-Kutta Fehlberg method of order Fehlberg a local error of order h^6 and general error of order h^5 which has higher accuracy than other methods. [13]

The designed program has a high ability to calculate the depletion of material in flux and constant power condition. To find the flux dependent time in constant power condition, we employed flux relation as follow:

Inverting Eq. (5), the flux as a function of time is written as: [5]

$$\phi = \frac{6.242 * 10^{18} P}{\sum_{i=1}^{N} X_i^f \sigma_i^f R_i} \qquad (5)$$

where P is the specific power in MW/cm3, X^P σ^f is the total macroscopic fission cross section of the fuel in cm^{-1}, and ϕ is the instantaneous value of the neutron flux in neutrons/cm2•s, R_i is released energy per fission reaction MeV/ fission that obtained from following Eq . [5]

$$R(MeV/fission) = 1.29927 * 10^{-3}(Z^2 A^{0.5}) + 33.12 \qquad (6)$$

Where, Z is the atomic number and A is mass number. Taylor expansion of the fluxat t = 0 is

$$\phi(t) = \phi(0) + t\dot{\phi}(0) + \frac{t^2}{2}\ddot{\phi}(0) + \frac{t^3}{2}\dddot{\phi}(0) + \ldots \qquad (7)$$

To obtain the average of the flux over the time interval, t, Eq. 6 is integrated over the interval (0,1) and divided by t.

$$\phi_{avr}(t) = \frac{1}{t}\int_0^t \phi(t')dt' = \phi(0) + \frac{t}{2}\dot{\phi}(0) + \frac{t^2}{6}\ddot{\phi}(0) + \frac{t^3}{24}\dddot{\phi}(0) + \ldots \qquad (8)$$

Where

$$\dot{\phi}(0) = -6.242*10^{18} P\left(\sum_{i=1}^{N} \dot{X}_i \sigma_i^f R_i\right)\left(\sum_{i=1}^{N} X_i \sigma_i^f R_i\right)^{-2} \qquad (9)$$

$$\ddot{\phi}(0) = 6.242*10^{18} P\left(-\left(\sum_{i=1}^{N} \ddot{X}_i(0) \sigma_i^f R_i\right)\left(\sum_{i=1}^{N} X_i(0) \sigma_i^f R_i\right)^{-2} + 2\left(\sum_{i=1}^{N} \dot{X}_i(0) \sigma_i^f R_i\right)^2\left(\sum_{i=1}^{N} X_i(0) \sigma_i^f R_i\right)^{-3}\right)$$

$$\dddot{\phi}(0) = 6.242*10^{18} P\left(-\left(\sum_{i=1}^{N} \dddot{X}_i(0) \sigma_i^f R_i\right)\left(\sum_{i=1}^{N} X_i(0) \sigma_i^f R_i\right)^{-2} + 6\left(\sum_{i=1}^{N} \dot{X}_i(0) \sigma_i^f R_i\right)\left(\sum_{i=1}^{N} \ddot{X}_i(0) \sigma_i^f R_i\right)\left(\sum_{i=1}^{N} X_i(0) \sigma_i^f R_i\right)^{-3} - 6\left(\sum_{i=1}^{N} \dot{X}_i(0) \sigma_i^f R_i\right)^3\left(\sum_{i=1}^{N} X_i(0) \sigma_i^f R_i\right)^{-4}\right)$$

The second and third derivative can also be expressed in terms of the nuclide concentrations as:

$$\ddot{X} = A\dot{X} = A^2 X \quad , \quad \dddot{X} = A\ddot{X} = A^2\dot{X} = A^3 X \qquad (10)$$

To evaluate the Equation.8 are used Eqs.9 and 10 that these equations provides the average flux as a vector that is sum of produced flux by various fission nuclides at the beginning of each time interval. In the computer program, the estimation for the average flux includes only the two terms of Equation (8) for all time intervals except the first them. At the first interval, the second derivative term will be significant because the first derivative term will be zero for many nuclides. [3, 4]

Results and discussions

In this paper, we extract decay constants and the reaction cross section of various elements from ORIGEN code different libraries. Therefore, we solved the problem by using the designed program to calculate the radioactive elements decay chain of differential equations. Also, we can calculate atomic density of the elements in the library for the initial conditions and the desired accuracy. In this section, the obtained results of the designed program to evaluate the calculation for Xe135 offcondition, with a density value about of 1.15×10^{15} and I^{135} with a density value about of 5×10^{15}, are shown in Figure.4.

Xenon-135 has thermal absorption cross section 2.6 *106 barns. It is produced directly from fission and from the decay of ^{135}I .

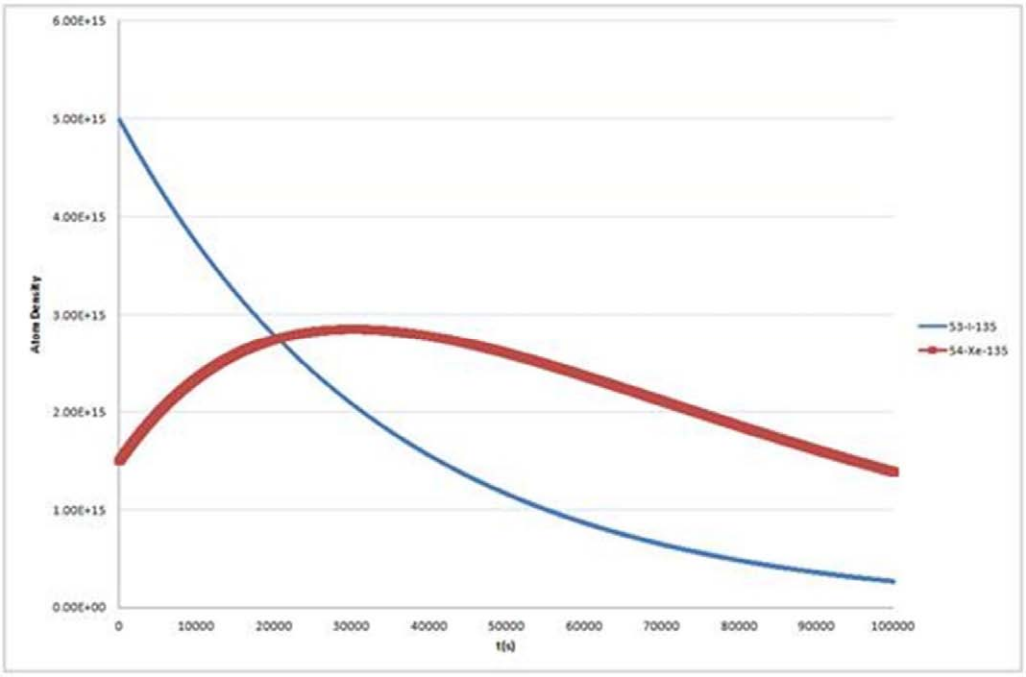

Figure 4. The rate of atomic density of "off Xe^{135}" in time for the reactor core, with a initial density value about of 1.15×10^{15} and I^{135} with a initial density value about of 5×10^{15} in designed program.

It should to be mentioned that by using the equation.11 and 12 as two analytical relations, with regard to the zero flux, we obtained the time depending of Xe^{135} and I^{135} changes.

$$I(t) = \frac{\gamma^{Te}\sum_f \phi}{\lambda^I}(1-e^{-\lambda_I t}) + I(0)e^{-\lambda_I t} \qquad (11)$$

$$X(t) = \frac{(\gamma^{Te}+\gamma^{Xe})\sum_f \phi}{\lambda^X + \sigma_a^X \phi}(1-e^{-(\lambda^X+\sigma_a^X\phi)t}) + \frac{\gamma^{Te}\sum_f \phi - \lambda^I I(0)}{\lambda^X + \sigma_a^X \phi - \lambda^I}(e^{-(\lambda^X+\sigma_a^X\phi)t} - e^{-\lambda^I t}) + X(0)e^{-(\lambda^X+\sigma_a^X\phi)t}$$

$$I(t) = I(0)e^{-\lambda^I t} = 5\times 10^{15} e^{-0.29\times 10^{-4} t} \qquad (12)$$

For example:

$$\text{If } t = 20291s \text{ then } I(t) = 5\times 10^{15} e^{-0.29\times 10^{-4}\times 20000} = 2.76e+15$$

Whereas, the atomic density of iodine at this time by the computational code is about of 2.74×10^{1} that the relative difference is about of 0.72%.

$$X(t) = \frac{-\lambda^{I} I(0)}{\lambda^{X} - \lambda^{I}}\left(e^{-\lambda^{X} t} - e^{-\lambda^{I} t}\right) + X(0)e^{-\lambda^{X} t}$$

$$= \frac{-0.29 \times 10^{-4} \times 5 \times 10^{15}}{0.212 \times 10^{-4} - 0.29 \times 10^{-4}}\left(e^{-0.212\times10^{-4} t} - e^{-0.29\times10^{-4} t}\right) + 1.5$$

$$\times 10^{15} e^{-0.212\times10^{-4} t} \qquad (9)$$

As second example:

If $t = 20291$ s then $X(t) = 2.75e + 15$

Whereas, the atomic density of iodine at this time by the computational code is about of 2.74e+15that the relative difference is about of 0.36%.

To evaluate more exact, we solved the decay chain of uranium238 in the constant flux is about of 5×10^{13} by analytical and Runge-Kutta Fehlberg methods that the obtained results are shown in Figure 5.

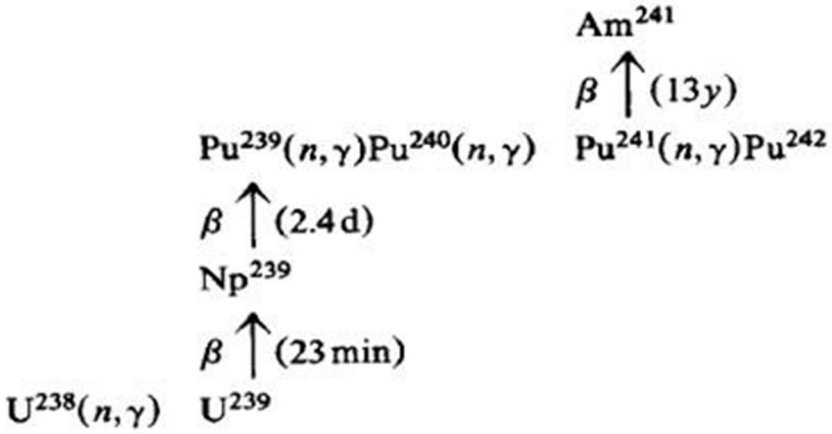

Figure 5. The schematic decay chain of uranium 238.

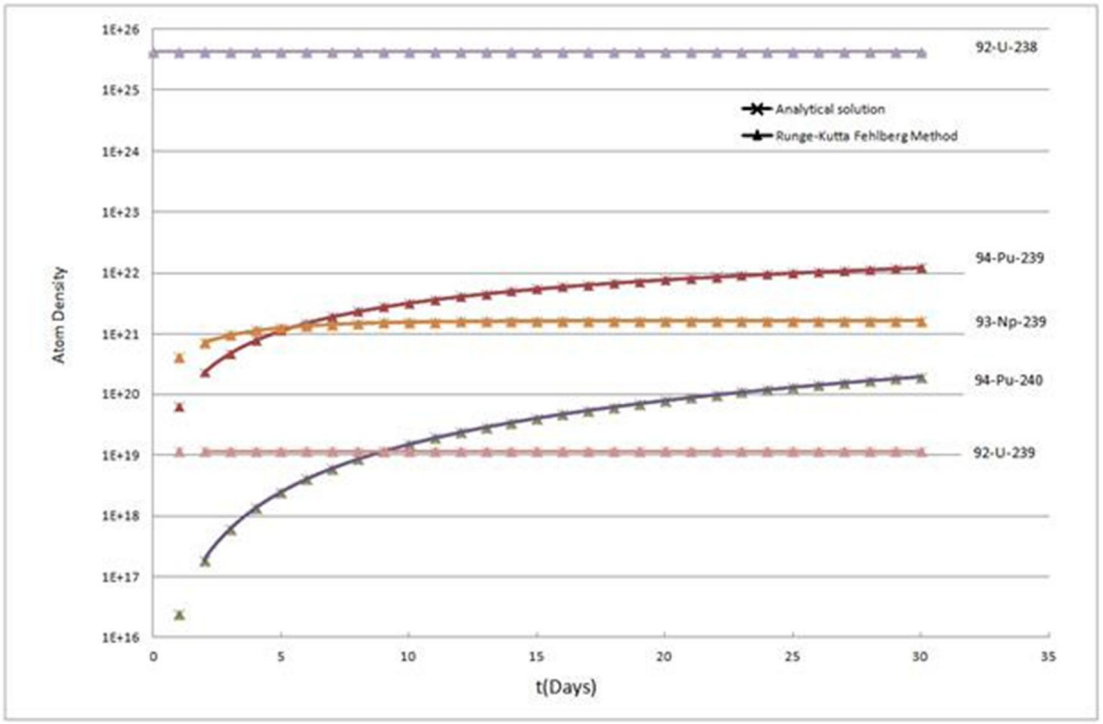

Figure 6. The obtained results of atomic density changes calculations for the decay chain of uranium-238.

We calculated the difference of density relative for nuclear material by analytical and Runge-Kutta Fehlberg methods for all point. The maximum difference relative is 0.176% and the least relative difference is 0.0068%. Changes in atomic density, uranium235, with the initial atomic density 4.2E24, with approximate constant power, 5MW, and constant flux, 6.3E13, calculated for the period of 100 days. The obtained results in the Figures 7 and 8 are shown.

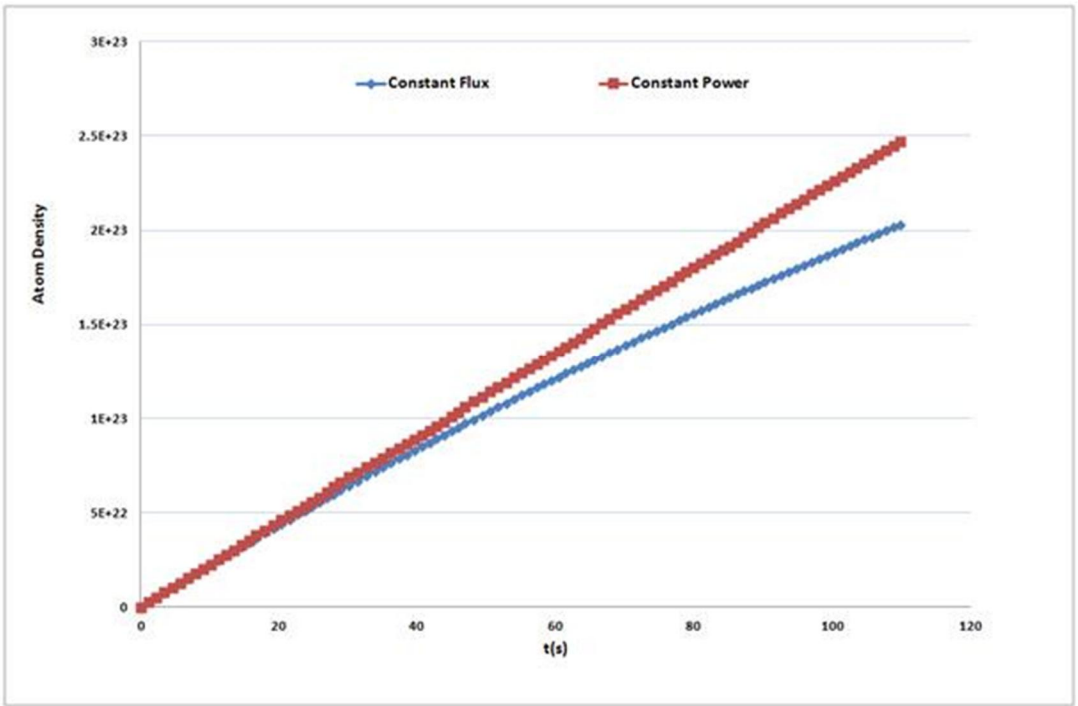

Figure 7. Atom density changes of uranium-236 in the constant flux or power approximation.

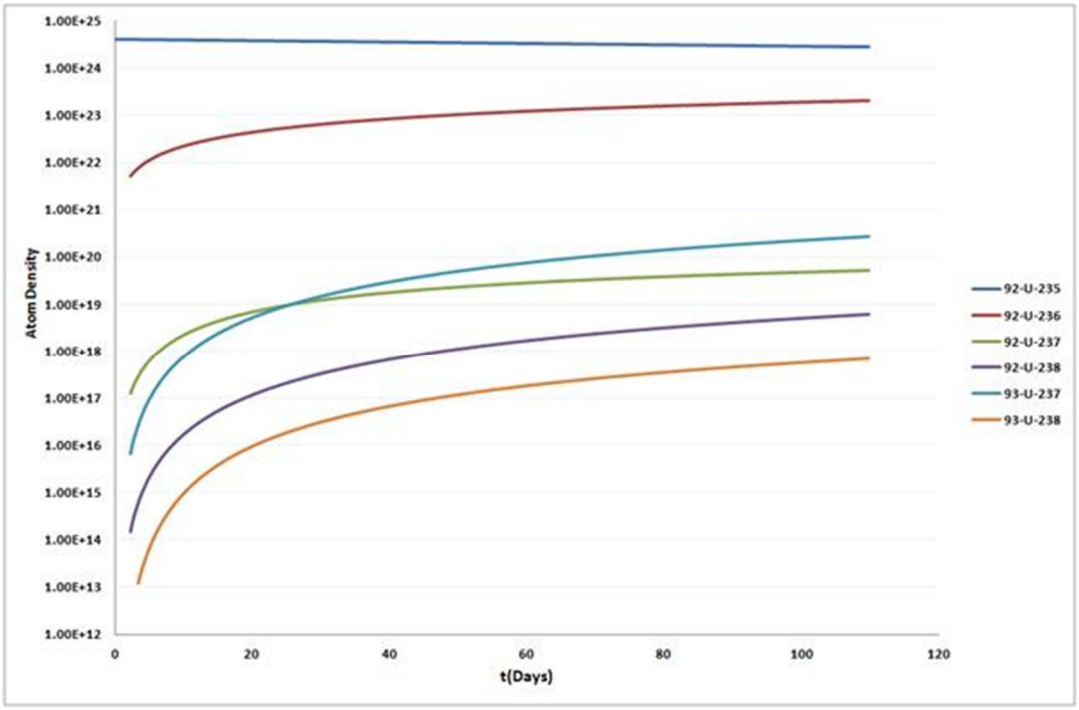

Figure 8. Atomic density changes of heavy nuclei that are produced by uranium-235 by using the approximate constant power and Runge-KuttaFehlberg method.

Conclusions

In this research, we designed the computational code by C# to solve the Bitman equations system by using Runge-Kutta Fehlberg method and approximations of flux, constant power and material power density calculation. The designed program is executed to different conditions that are fully explained in last section. The obtained results of this method are compared to the obtained results of analytical solutions for these conditions. These results showed a small difference between analytical solutions and Runge-Kutta Fehlberg method that are represented adequate accuracy of Runge-Kutta Fehlberg method by setting network pitch.

Finally, we achieved to the computational codeto calculate the 1700 atomic density changes in the reactor operations with the high ability to calculate the material depletion with constant flux and constant power condition.

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References

[1] W.M. Stacey, *Nuclear Reactor Physics*, John Wiley and Sons (2007).

[2] J. J. Duderstadt, J. Louis, J. Hamilton, *Nuclear Reactor Analysis*, Wiley and Sons (1976).

[3] C.K. Gupta, *Science and Technology*, 63, 39

(2001).

[4] M. T. Simnad, *Science and Technology* (Third Edition), 775 (2003).

[5] A.G. Croff, "*A User's Manual forthe ORIGEN2 computer code,"* ORNL/TM-7175 (CCC-371), Oak Ridge National Laboratory (1980).

[6] R. M .W Hermann, *Scale system module to calculate fuel depletion, actinide transmutation, fission product buildup and decay, and associated radiation source terms* (1998).

[7] K. Kikuchi, *Comprehensive Nuclear Materials*, 5, 207 (2012).

[8] P. Yvon, F. Carré, *Journal of Nuclear Materials*, 385, 217 (2009)

[9] P. Pareige, R.E. Stoller, K.F. Russell, M.K. Miller, *Journal of Nuclear Materials*, 249, 165 (1997)

[10] H.U. Zwicky, J. Low, M. Granfors, C. Alejano, J.M. Conde, C. Casado, J. Sabater, M. Lloret, M. Quecedo, *Journal of Nuclear Materials*, 402, 60 (2010).

[11] A.E. Aboanber, *Annals of Nuclear Energy*, 28, 933 (2001).

[12] A. G .Croff, *Nuclear Tech*., 62, 335 (1983).

[13] S. Ludwig, ORIGEN2, Version 2.1 (8-1-91) Released Notes (1999).

[14] http://math-cs.aut.ac.ir/~shamsi/Ebooks/Kreyszig/Ch21%20from%20Kreyszig%20e-24.pdf.

[15] http://www.pit.physik.uni-tuebingen.de/~speith/Projekt1/numrec/c16-2.pdf.