JPPIPA 7(3) (2021)

Jurnal Penelitian Pendidikan IPA

Journal of Research in Science Education

http://jppipa.unram.ac.id/index.php/jppipa/index

Numerical Solution of Radioactive Core Decay Activity Rate of Actinium Series Using Matrix Algebra Method

Anita Puji Pratiwi[1*], Trapsilo Prihandono[1], Sri Handono Budi Prastowo[1]

[1] Department of Physics Education, University of Jember, Jember, Indonesia

DOI: 10.29303/jppipa.v7i3.716

Article Info
Received: April 20th, 2021
Revised: July 2th, 2021
Accepted: July 7th, 2021

Abstract: The Actinium 235 series is one of the radioactive series which is widely used as a raw material for reactors and nuclear activities. The existence of this series is found in several countries such as West USA, Canada, Australia, South Africa, Russia, and Zaire. The purpose of this study was to determine the activity value and the number of radioactive nucleus decay atoms on the actinium 235 rendered in a very long decay time of 4.3 x 10^9 years. The decay count in this study uses an algebraic matrix method to simplify the chain decay solution, which generally uses the concept of differential equations. The solution using this method can be computationally simulated using the Matlab program. This study indicates that the value of the decay activity experienced by each element in this series is the same, which is equal to 2,636 x 10^{11} Bq. This condition causes the actinium 235 series to experience secular equilibrium because the half-life of the parent nuclide is greater than the nuclide derivatives. The decay activity of the radioactive nucleus under the actinium 235 series is strongly influenced by the half-life of the nuclides, the decay constants, and the number of atoms after decay.

Keywords: Actinium 235 Series; Chain Decay; Decay Activity; Matrical Algebra Method.

Citation: Pratiwi, A., Prihandono, T., & Prastowo, S. (2021). Numerical Solution of Radioactive Core Decay Activity Rate of Actinium Series Using Matrix Algebra Method. *Jurnal Penelitian Pendidikan IPA, 7*(3), 395-400. doi:https://doi.org/10.29303/jppipa.v7i3.716

Introduction

The actinium series is one of the natural radionuclide series formed from the decay process of the radioactive element Uranium-238. This series is one of the types of elements that still exist in nature, even though the amount is not as much as Thorium and Uranium's elements. The decay stage of this series starts with Actinium-235 and ends at the stable actinium, namely Timbal-207. The parent of the nuclides in the Actinium series has a half-life of 7.10 x 10^8 years (Sofyan *et al.*, 2004). The Actinium 235 series is used as raw material in Nuclear and actinium installations. The fuel is part of the uranium series, where the production of this fuel comes from pure natural uranium (*Uranite*), which is processed into *yellowcake*. Uranium ore *(Yellowcake)*itself contains natural uranium contained pure isotope U-234, U-235, and U-238. The required content of U-235 is 0.72% and is enlarged again to 3% to be used on the HWR actinium. These uranium seeds are found in the Western USA, Canada, Australia, South Africa, Russia, and Zaire, which are also used as fuel for actinium and nuclear activities (Alatas *et al.*, 2015).

The radionuclide series in the form of Actinium 235 series was found in all environmental media, one of which was on the coast of Bengkalis Island. The radinuclide activity of the actinium series in the area has an average value that is still within reasonable limits (Makmur., 2019). Noviarty (2017) also adds that the radioactive activity of the actinium U-235 series ranges from 0.013 Bq / g found in rocks around Batan

Email: pratiwianita387@gmail.com

in Banten, so the danger posed is not too heavy. This radioactive activity is the decay of the nucleus which converts unstable nuclides into stable nuclides, hereinafter referred to as the radionuclide core decay rate. The decay of the nuclide decay activity (*R*) is defined as the amount of *N* decay (Sign (-) indicates that *N* decreases with time) that occurs per unit of time, so (*R*) can also be written in the following form:

$$R = \frac{dN}{dt} = -\lambda N \quad (1)$$

$$\frac{dN}{N} = -\lambda\, dt \quad (2)$$

$$\int_{N_{(0)}}^{N_{(t)}} \frac{dN}{N} = -\lambda \int_{0}^{t} dt \quad (3)$$

$$\ln N_{(t)} - \ln N_0 = -\lambda t \quad (4)$$

$$N_t = N_0 e^{-\lambda t} \quad (5)$$

where N_t is the number of atoms of decay, N_0 is the initial number of atoms, λ is the decay constant, and t is the time of decay.

The decay activity (R) of the Actinium 235 series continues to decrease with time. The rate at which a radioactive substance decays to the end is determined by the half-life ($t_{1/2}$) of the nuclide. Based on previous research by Suparjo (2014: 39) the amount of decay depends on the number of initial atoms (N_0) and the disentegration constant (λ) which depends on the length of half-life ($t_{1/2}$) that each nuclide has. The longer the half-life of the nuclides, the greater the number of atoms resulting from the decay of the nuclides. The following is a table of half-lives for the Actinium 235 series:

Table 1. The half-life of the Actinium 235 series

Decay to	uclide	Symbol	Half-life (t1/2)
1.	Actinouranium	$^{235}_{92}U$	7,10 x 10^8 years
2.	Uranium Y	$^{231}_{90}Th$	25,6 hours
3.	Protactinium	$^{231}_{91}Pa$	3,43 x 10^4 years
4.	Actinium	$^{227}_{89}Ac$	21,6 years
5.	Radioactinium	$^{227}_{90}Th$	18,17 days
6.	Actinium X	$^{223}_{88}Ra$	11,68 days
7.	Ac Emanation	$^{219}_{86}Rn$	3,92 seconds
8.	Actinium A	$^{215}_{84}Po$	1,83 x 10^{-3} seconds
9.	Actinium B	$^{211}_{82}Pb$	36,1 minutes
10.	Actinium C	$^{211}_{83}Bi$	2,15 minutes
11.	Actinium C	$^{211}_{84}Po$	0,52 seconds
12.	Actinium D	$^{207}_{82}Pb$	Stable

Sofyan, (2004)

The actinium 235 series undergo 11 decay processes consisting of 7 alpha (α) and 4 beta (β) decays. As for the decay process, it produces an atomic nucleus with mass number divided by 4 by the remainder of 3. In brief, the decay process of the actinium 235 series can be written as follows:

$$^{235}_{92}U \longrightarrow {}^{207}_{82}Pb + 7\,{}^{4}_{2}\alpha + 4\,{}^{0}_{+1}\beta$$

The chain decay that occurs in the actinium 235 series can be solved using the Bateman equation, (Ridwan et al., 2015: 177). This equation develops linear equations into differential and integral equations, Cetnar (2006: 640). The following forms a linear equation which is developed into a differential equation

$$N_1(t) = -\lambda_1 N_1 \longrightarrow \frac{dN_1}{dt} = -\lambda_1 N_1 \quad (6)$$

$$N_2(t) = \lambda_1 N_1 - \lambda_2 N_2 \longrightarrow \frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \quad (7)$$

$$N_3(t) = \lambda_2 N_2 - \lambda_3 N_3 \longrightarrow \frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3 \quad (8)$$

Yuan (2007:1) states that Bateman's equation uses the trans Laplace form to solve differential equations. In fact, the use of this method still has its drawbacks even though it has been used for many years. The weakness is that this equation is still imperfect because it requires a very complicated counting process for long decay.

Based on the description above, a counting method is needed regarding the rate of decay activity in the actinium 235 series using an easier method. Therefore, a research entitled "Numerical Solutions for the Activity Rate of Radioactive Core Decay of Actinium Series Using the Matrix Algebra Method" needs to be done as a solution to the above problems. This research was conducted using a matrix algebra method which examines the processes that take place from the formation of the earth until the series can achieve stability. In addition, this research was conducted in order to determine the rate of decay activity that occurs in the actinium 235 series numerically using the matrix algebra method.

Method

The research method used is a matrix algebraic method centered on chain decay in the actinium 235 series. Previously we mentioned that the solution to the chain decay count applies the Bateman equation system. Therefore, the mathematical algebra method in this study is a method that applies the Bateman equation system in a way that develops linear equation models into differential and integral equations. Through this method, a numerical solution of

differential equations in chain decay is obtained which requires a long process.

A solution to solving a chain decay can be written using a matrix form as follows:

$$\boldsymbol{N' = AN} \tag{9}$$

Based on the research of Yushardi et al. (2020: 3), the N' component is the number of nuclei as a result of the decay and N is the initial number of nuclei, and A is the decay constant if expressed into the matrix, it will be :

$$\begin{bmatrix} N_1' \\ N_2' \\ N_3' \\ \vdots \\ N_n' \end{bmatrix} = \begin{bmatrix} -\lambda_1 & 0 & 0 & \dots & 0 \\ \lambda_1 & -\lambda_2 & 0 & \dots & 0 \\ 0 & \lambda_2 & -\lambda_3 & \dots & 0 \\ \dots & \dots & \ddots & \ddots & \dots \\ 0 & 0 & 0 & \lambda_{n-1} & -\lambda_n \end{bmatrix} \text{x} \begin{bmatrix} N_1 \\ N_2 \\ N_3 \\ \vdots \\ N_n \end{bmatrix} \tag{10}$$

Based on Taylor's exponential equation, the form of equation (5) can be analogized to the following form:

$$N(t) = e^{At} N_0 \tag{11}$$

Solving the chain decay event with the exponential matrix equation form e^{At} can use eigenvalues. The resulting form of solving equation (11) using the eigenvalues will be in the form below:

$$\boldsymbol{N(t) = e^{VDV^{-1}t} N_0} \tag{12}$$

Furthermore, to get the real matrix form, we can use the Taylor exponential expansion concept to be analogized to a matrix form. The final result of this method in the form of a serial equation solution in the form of a matrix as below:

$$\boldsymbol{N(t) = V\, e^{Dt}\, V^{-1}\, N_0} \tag{13}$$

where N_t is the number of remaining atoms of decay during time t, $\boldsymbol{V}$ is the decay constant eigenvector matrix, $\boldsymbol{V^{-1}}$ is the inverse matrix of the decay constant eigenvectors. The $\boldsymbol{e^{Dt}}$ matrix is a type of negative exponential diagonal matrix of the decay constant as shown below:

$$e^{Dt} = \begin{bmatrix} e^{-\lambda_1 t} & 0 & 0 & \dots & 0 \\ 0 & e^{-\lambda_2 t} & 0 & \dots & 0 \\ 0 & 0 & e^{-\lambda_3 t} & \dots & 0 \\ \dots & \dots & \ddots & \ddots & \dots \\ 0 & 0 & 0 & 0 & e^{-\lambda_n t} \end{bmatrix} \tag{14}$$

The use of eigenvalues theory will make it easier to find the components of the matrix $\boldsymbol{V}$ and $\boldsymbol{V^{-1}}$. Levy (2018: 911) states that the matrix $\boldsymbol{V}$ form is a type of lower triangle matrix, with the principle that $S_{ij} = 0$ if $i < j$ and $S_{ij} = 1$ if $i = j$, (i =rows and j = columns)

$$V = \begin{bmatrix} 1 & 0 & 0 & \dots & 0 \\ S_{2,1} & 1 & 0 & \dots & 0 \\ S_{3,1} & S_{3,2} & 1 & \dots & 0 \\ \dots & \dots & \ddots & \ddots & \dots \\ S_{n,1} & S_{n,2} & S_{n,3} & S_{n,n-1} & 1 \end{bmatrix} \tag{15}$$

Where the components $\boldsymbol{S_{2,1}}$, $\boldsymbol{S_{3,1}}$ to $\boldsymbol{S_{n,1}}$ are the partial decay constants of the first nuclide (λ_1), while $\boldsymbol{S_{3,2}}$ is the partial decay constant of the second nuclide (λ_2).

The component of the matrix $\boldsymbol{V^{-1}}$ is obtained by inversing matrix (14) so that it is obtained:

$$V^{-1} = \begin{bmatrix} 1 & 0 & 0 & \dots & 0 \\ T_{2,1} & 1 & 0 & \dots & 0 \\ T_{3,1} & T_{3,2} & 1 & \dots & 0 \\ \dots & \dots & \ddots & \ddots & \dots \\ T_{n,1} & T_{n,2} & T_{n,3} & T_{n,n-1} & 1 \end{bmatrix} \tag{16}$$

The analysis of the components in matrices (14) and (15) is represented as a product like the following:

$$F_{q,r}^{p} = \frac{\lambda_r}{\lambda_q - \lambda_p} \tag{17}$$

The values of $S_{x,y}$ dan $T_{x,y}$ adalah:

$$S_{x,y} = F_{x,x-1}^{y} F_{x-1,x-2}^{y} \dots F_{y+1,y}^{y} \tag{18}$$

$$T_{x,y} = F_{y,y}^{x} F_{y+1,y+1}^{x} \dots F_{x-1,x-1}^{x} \tag{19}$$

Furthermore, equations (14), (15) and (16) can be substituted for equation (13), so that the final matrix form becomes the numerical solution for the decay of Actinium 235:

$$\begin{bmatrix} N_1' \\ N_2' \\ N_3' \end{bmatrix} = \begin{bmatrix} 1 & 0 & 0 \\ S_{2,1} & 1 & 0 \\ S_{3,1} & S_{3,2} & 1 \end{bmatrix} \text{x} \begin{bmatrix} e^{-\lambda_1 t} & 0 & 0 \\ 0 & e^{-\lambda_2 t} & 0 \\ 0 & 0 & e^{-\lambda_3 t} \end{bmatrix} \text{x} \begin{bmatrix} 1 & 0 & 0 \\ T_{2,1} & 1 & 0 \\ T_{3,1} & T_{3,2} & 1 \end{bmatrix} \text{x} \begin{bmatrix} N_1(0) \\ 0 \\ 0 \end{bmatrix} \tag{20}$$

This research on serial decay of the Actinium 235 series was carried out in the even semester of the 2021-2022 academic year with a place in the Advanced Physics Laboratory, Physics Education, Faculty of Teacher Training and Education, University of Jember. The stages of this research are preparation, theoretical study, simulation, validation, discussion, and conclusion. Simulations carried out using the Matlab program with some inputted data include the decay time (t) for 4.3 x 10^9 years, the half-life of each nuclide

$t_{1/2}$ mass 7 grams, avogadro number. The data presented will be in the form of a research table as well as a graphic visualization of the decay process. The results of the simulation were analyzed descriptively regarding the decay activity rate of the Actinium 235 series.

Result and Discussion

The decay of the Actinium 235 series took place from the time the earth was formed, which was 4.3 x 10^9 years. By using a mass of 7 grams, the initial number of atoms N_0 is 1.79 x 10^{22} atoms. This initial atomic number is used as the initial variable to calculate the number of remaining atoms and the decay activity experienced by each nuclide in the Actinium 235 series. The process of decaying the Actinium 235 series using the Matlab program produces the following data :

Table 2. Data on the Simulation of Decay of Actinium 235 Series Decay

No.	Nuclide	Symbol	N_t (atom)	R (Bq)
1	Actinouranium	$^{235}_{92}U$	2,695 x 10^{20}	3,649 x 10^3
2	Uranium Y	$^{231}_{90}Th$	1,109 x 10^9	3,649 x 10^3
3	Protactinium	$^{231}_{91}Pa$	1,302 x 10^{16}	3,649 x 10^3
4	Actinium	$^{227}_{89}Ac$	8,201 x 10^{12}	3,649 x 10^3
5	Radioactinium	$^{227}_{90}Th$	1,890 x 10^{10}	3,649 x 10^3
6	Actinium X	$^{223}_{88}Ra$	1,215 x 10^{10}	3,649 x 10^3
7	Ac Emanation	$^{219}_{86}Rn$	4,719 x 10^4	3,649 x 10^3
8	Actinium A	$^{215}_{84}Po$	2,203 x 10^1	3,649 x 10^3
9	Actinium B	$^{211}_{82}Pb$	2,608 x 10^7	3,649 x 10^3
10	Actinium C	$^{211}_{83}Bi$	1,553 x 10^6	3,649 x 10^3
11	Actinium C'	$^{211}_{84}Po$	6,260 x 10^3	3,649 x 10^3
12	Actinium D	$^{207}_{82}Pb$	2,632 x 10^{11}	3,649 x 10^3

Based on the table above, it can be stated that the largest number of decay atoms experienced by the parent nuclide, namely U-235 of 2,695 x 10^{20} atoms. At the same time, the smallest number of atoms occurs in the daughter nuclides, namely Po-215 of 2.203 x 10^1 atoms. The difference results from the number of decay atoms experienced by the nuclides U-235 and Po-215 due to the difference in half-lives. The half-life of the parent nuclide U-235 is 7.10 x 10^8 years (the largest half-life in the actinium 235 series), while the half-life of the nuclides Po-215 is the smallest is 1.83 x 10^{-3} seconds. Therefore, it is stated that the greater the half-life, the greater the number of decay atoms.

Meanwhile, another factor that affects the number of atoms resulting from the decay is the decay constant (λ). The largest decay constant value is 1.19 x $10^{10,}$ which is the child nuclide Po-215. Whereas the smallest decay constant occurs in the parent nuclide U-235 with 9.76 x 10^{-10}. Therefore, it can be concluded that the decay constant (λ) is inversely proportional to its half-time value ($t_{1/2}$). This means that the smaller the half-life of a nuclide, the greater the value of its decay constant. As a result, this will make the number of atoms decay (N_t) smaller, and vice versa.

The value of the decay activity in the Actinium 235 series is the same for all radionuclides in the Actinium 235 series, which is 3.649 x 10^3 Bq. Therefore, it can be stated that the decay experienced by the Actinium series 235 reaches its stability with a large decay activity of 3.649 x 10^3 Bq. The decay process that occurs in this series takes 1.356 x 10^{18} seconds or the equivalent of 4.3 x 10^{13} years to reach radioactive equilibrium. This is also clearly seen in the graphic image below, where this series of Actinium 235 achieve an equilibrium coordinate point.

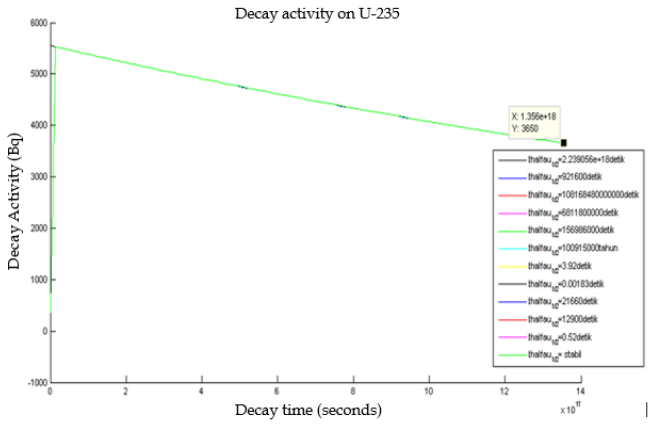

Figure 1. Graph of the Relationship between Decay Activity and Decay Time

Figure 1. illustrates the decay activity of the parent nuclides of the actinium 235 series when a time of 1.356 x 1018 seconds is the same as the nuclide derivatives. When the value of the decay activity of all nuclides in this series is the same, this condition indicates that the actinium 235 series is in secular equilibrium. This condition is a condition where the decay activity rate of the parent nuclide will be in the same condition as the nuclide derivatives. The factor that makes the secular equilibrium occur is the parent nuclides' half-life value, which is much greater than the half-life of its child nuclides ($t_{1/2(Induk)} \gg t_{1/2(Anak)}$). Another factor is the value of the parent nuclide constant, which is very small compared to the child nuclide constant ($\lambda_{Induk} \ll \lambda_{Anak}$). This is in great agreement with the decay results in this series.

The secular equilibrium that occurs in the actinium 235 series also causes the formation of the most stable derivative nuclide, namely Pb-207, when the parent nuclide decays until it reaches stability. In addition, the number of atoms of decay also affects the value of the decay activity. It is known that the number

of decay atoms per nuclide represents a reduced decay activity due to the decreasing number of N atoms with time. This is in accordance with Graph 1. which depicts decay activity decreasing in a certain time (seconds). This decay activity stops at a point of stability where the parent atom has decayed to become a stable nuclide (Pb-207).

The advantage of using this matrix algebra method is that it has a simple form of solution and is easy to apply to computational systems. This condition is similar to what has been mentioned by Yuan and Kernan (2007), compared to using the sequential substitution method or the Laplace transform method, resulting in complex calculations. Meanwhile, the weakness of using this method is the long and repetitive calculation process, so that high accuracy is required. This condition is caused by the decay that occurs in the radionuclide series in the Actinium 235 series, which requires a very long process to achieve stability.

Conclusion

Based on the data analysis previously described, several conclusions were obtained as follows: (1) The numerical solution of the number of decay atoms in the Actinium 235 series explains that the highest number of decay atoms occurs in the parent nuclide, which is 2,695 x 10^{20} atoms. The number of atoms of this decay is influenced by the number of atoms before decay and the decay constant (λ). The greater the decay constant (λ), the smaller the number of decay atoms produced. The value of the decay constant is inversely proportional to the half-life of each nuclide. Therefore the number of decay atoms will increase in number if the half-life is long; (2) The numerical solution of the actinium 235 series decay activity shows that the decay activity has the same value, which is 3.649 x 103 Bq. The value of the decay activity is achieved after going through an interval of 1.356 x 10^{18} seconds or equal to 4.3 x 10^{13} years. The value of this decay activity is strongly influenced by the number of atoms after decay and the decay constant (λ). The same amount of decay activity causes a secular equilibrium, which is a condition caused by the larger half-life of the parent nuclide compared to the nuclide derivatives. This event causes the creation of stable daughter nuclides at the same time that the parent nuclides decay.

Based on the research entitled "Numerical Solutions for the Radioactive Core Decay Activity Rate of the Actinium Series Using the Matrix Algebra Method," it is hoped that readers will be able to use it as a reference in the discussion of the decay of the atomic nuclei of radioactive substances. In addition, it is hoped that this research can be used as a comparison to similar studies. Furthermore, through this research, it is hoped that it can also be an inspiration for readers or other researchers in the future to develop the discussion described above by using different methods or variables.

Acknowledgments

The research journal entitled "Numerical Solutions for the Core Radioactive Decay Activity Rate of Actinium Series Using the Matrix Algebra Method" expresses sincere gratitude to my mother and father, who always provide constructive advice and motivation, as well as colleagues in arms who never stop supporting me in the process of implementing this research

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