CALCULATION OF RADIONUCLIDE CONTENT OF NUCLEAR MATERIALS USING ORIGEN2.1 COMPUTER CODE

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ABSTRACT

CALCULATION OF RADIONUCLIDE CONTENT OF NUCLEAR MATERIALS USING ORIGEN2.1 COMPUTER CODE. Nuclear materials contain a number of radionuclides produced from radioactive decay process. The composition of these radionuclides which are accumulated inside the nuclear materials changes over the time. The calculation of radionuclide composition inside nuclear materials is very important especially in the aspect of nuclear reactor safety evaluation, nuclear fuel behavior evaluation, and radioactive waste management. One method to calculate radionuclide content of nuclear materials is by using ORIGEN2.1 computer code. Beside radionuclide composition, this code can also calculate some characteristics related to decay process such as total radioactivity, decay heat, and neutron flux. This paper is a literature study about ORIGEN2.1 computer code. A brief description of ORIGEN2.1 and its use for calculating radionuclide content of nuclear materials are presented. Radionuclide content produced from californium-252 decay was chosen as a simple case solved by ORIGEN2.1. Californium-252 was simulated to undergo decay for 10 years. The variables which are calculated by ORIGEN2.1 in this case are radionuclide composition, total radioactivity, total alpha radioactivity, and neutron flux. From the results of this simulation, it is shown that small amount of californium-252 produces high neutron intensity so that it can be used as a reliable neutron source for many applications.

Keywords: ORIGEN2.1, radioactive decay, radionuclide content, californium-252

ABSTRAK

PERHITUNGAN KANDUNGAN RADIONUKLIDA MATERIAL NUKLIR DENGAN MENGGUNAKAN KODE KOMPUTER ORIGEN2.1. *Suatu material nuklir yang di dalamnya terdapat unsur-unsur radioaktif mengandung sejumlah radionuklida hasil peluruhan. Radionuklida-radionuklida hasil perluruhan ini terakumulasi di dalam material nuklir dengan komposisi yang selalu berubah setiap waktu. Perhitungan komposisi radionuklida di dalam suatu material nuklir sangat penting untuk diketahui terutama terkait evaluasi keselamatan reaktor nuklir, manajemen bahan bakar reaktor nuklir, dan manajemen limbah radioaktif. Salah satu metode untuk menghitung kandungan radionuklida material nuklir adalah dengan menggunakan kode komputer ORIGEN2.1. Selain komposisi radionuklida, ORIGEN2.1 juga bisa menghitung beberapa karakteristik yang berkaitan dengan proses peluruhan seperti radioaktivitas total, panas peluruhan, serta fluks neutron Makalah ini merupakan hasil studi literatur tentang kode komputer ORIGEN2.1. Deskripsi umum serta penggunaan ORIGEN2.1 untuk menghitung kandungan radionuklida dijelaskan secara singkat di dalam makalah ini. Sebagai contoh penggunaan ORIGEN2.1 dipilih suatu kasus sederhana yaitu peluruhan Californium-252. Californium-252 disimulasikan mengalami peluruhan selama 10 tahun. Besaran yang dihitung dengan ORIGEN2.1 pada kasus ini adalah komposisi radionuklida, radioaktivitas total, radioaktivitas partikel alfa total, serta fluks neutron. Dari hasil simulasi ini bisa disimpulkan bahwa satu gram californium-252 menghasilkan neutron dengan intensitas tinggi sehingga californium-252 dapat digunakan sebagai sumber neutron pada berbagai aplikasi.*

Kata kunci *: ORIGEN2.1, peluruhan radioaktif, kandungan radionuklida, californium-252*

INTRODUCTION

Radioactive decay is a transmutation process of an unstable nuclide into another nuclide by releasing certain radiation. The decay process occurs gradually until a stable nuclide is formed. Some radionuclides decay in several steps until reaching the formation of a stable nuclide. This type of decay is called decay chain. Nuclear materials in which decay chain occurs contain a number of nuclide daughters produced in each step of the decay processes. Nuclide daughters are often also radioactive so that they will undergo further decay and produce other nuclide daughters. The formation and decay process of the radionuclides make the composition of the radionuclide content inside a nuclear material changes as a function of time.

Nuclear fuels inside the nuclear reactor core are an example of a nuclear material, which contains a number of radionuclides. These radionuclides are mostly fission products produced from fission reactions inside the fuels. Beside fission products, there are also radionuclides produced from decay processes of each fission products and from neutron capture reactions of other materials in the reactor core. In addition, nuclear spent fuels still contain a number of radionuclides although there is no longer fission reaction occurring within it. These radionuclides are originated from the decay chain of the remaining radionuclides inside the spent fuels. The composition of the radionuclide content of the nuclear spent fuels also changes as a function of time [1]. In regard to nuclear fuel management, it is important to determine the composition of the radionuclide inventory inside the fuels in the reactor core. For example, some fission products, such as xenon and samarium, are neutron poison meaning that they absorb neutron and then change the reactor criticality. The amounts of these two radionuclides have to be calculated in order to decide the way to overcome this condition.

In the case of reactor safety evaluation, the number of radionuclides released from the reactor core into the environment has to be determined to make sure that the radioactivity level around the nuclear reactor are safe for the society. Moreover, in the case of nuclear reactor accident, the calculation of radionuclides released to the environment is very important to identify the severity of the accident and to determine the appropriate action and mitigation to be taken. The calculation of radionuclide content inside nuclear spent fuels are also important to quantify the radionuclide composition as well as its radioactivity and decay heat so that it leads to the appropriate way in handling and storing the spent fuels .

The most widely used method to calculate radionuclide content of nuclear materials is by using ORIGEN computer code. This code was developed by Oak Ridge National Laboratory (ORNL), USA. Beside radionuclide composition, ORIGEN code can also calculate the radioactivity of each radionuclide, decay heat, and some other characteristics of nuclear materials. This paper is a literature study about ORIGEN computer code. A brief description of ORIGEN code and its use in several cases are presented in this paper. For a study purpose, one simple decay case of Californium-252 was simulated. The calculations were done by using ORIGEN version 2.1. The variables which are computed by ORIGEN2.1 in this case are the composition of radionuclides along with its radioactivity, alpha particle radioactivity, and neutron fluxes.

A BRIEF DESCRIPTION OF ORIGEN

ORIGEN (Oak Ridge Isotope GENeration) is a computer code for calculating the buildup, decay, and processing of radioactive materials developed by Oak Ridge National Laboratory (ORNL) in the early 1970s. ORIGEN is originally developed to calculate the characteristics of nuclear spent fuels and radioactive wastes (nuclides composition, decay heat, and etc.). The calculation base of ORIGEN is solving the first-order linear differential equation for buildup and decay of radionuclides [2], which is shown as below:

$$\frac{dX_i}{dt} = \sum_{j=1}^{N} l_{ij}\lambda_j X_j + \varphi \sum_{k=1}^{N} f_{ik}\sigma_k X_k - (\lambda_i + \varphi\sigma_i + r_i)X_i + F_i, \quad i = 1, \ldots, N$$

where

X_i	= density of nuclide i
N	= number of nuclides
l_{ij}	= fraction of radioactive disintegration by other nuclide, which lead to formation of species i
φ	= position- and energy-averaged neutron flux
f_{ik}	= fraction of neutron absorption by other nuclides, which lead to formation of species i
σ_k	= spectrum-averaged neutron absorption cross section of nuclide k
r_i	= continuous removal rate of nuclide i from the system
F_i	= continuous feed rate of nuclide i

In several years later, the ORNL team took effort to update the original version of ORIGEN and launched ORIGEN2 in 1980s. In 1991, ORIGEN version 2.1 was launched. The data bases which are updated in ORIGEN2.1 are reactor models, cross section data, fission product yields, decay data, and decay photon data. The data bases of ORIGEN2.1 are divided into three segments: 130 actinides, 850 fission products, and 720 activation products. For each segment, there are three library data which are decay data, fission product yields, and photon decay data. ORIGEN2.1 is written in FORTRAN language and uses several input and output units to facilitate the operational of program become more flexible [3].

The characteristics of nuclear materials computed by ORIGEN2.1 are mass, radioactivity, thermal power, neutron emission, photon emission and etc. In case of nuclear reactor fuel cycle simulation, ORIGEN2.1 data bases are available for Liquid Water Reactor (LWR), Liquid Metal Fast Breeder Reactor (LMFBR), Molten Salt Breeder Reactor (MSBR), and High Temperature Gas-Cooled Reactor (HTGR). For HTGR, the library data have to be modified because ORIGEN2.1 has no cross section library data for high temperature reactor [4,5]. Library data modification is also needed when ORIGEN2.1 is used for simulating fuel cycle of research reactor [6,7].

The most widely use of ORIGEN2.1 is for calculating radionuclide inventory in the nuclear reactor core. The calculation of radionuclides inventory in the normal operation or in accident condition of nuclear reactor can be used to determine how many radionuclides released to the environment[8]. Moreover, the ORIGEN2.1 simulation can be used in some research cases such as nuclear accident analysis [9], nuclear fuel evaluation [10,11], and spent fuel analysis [12].

METHODOLOGY

This paper is a literature study about ORIGEN computer code focusing on ORIGEN 2.1. For a study purpose, decay of californium-252 was simulated by using ORIGEN2.1 in order to get an understanding about how to prepare the input file for ORIGEN2.1 and how to read and analyze the output data. Californium-252 (Cf-252) is an artificial radionuclide with half-life 2.6 years and decays in two modes: alpha emission (96.91 %) and spontaneous fission reaction (3.09 %). Cf-252 is known as a versatile radioisotope due to its wide application in many applications such as medicine, scientific research, industry, and nuclear science education.

Cf-252 is a unique neutron source which can provide a highly concentrated flux neutron from a very small assembly. This makes Cf-252 as a reliable neutron source. Cf-252 has been utilized and given a great success to cancer therapy, neutron radiography, neutron activation analysis, startup source for nuclear reactors, nuclear fuel analysis, and many others [13].

In this study, one milligram Cf-252 was assumed to undergo decay for 10 years. Some variables, which are radionuclide composition along with its radioactivity, alpha particle radioactivity, and neutron flux are calculated in several time intervals between 0.1 year and 10 years.

RESULTS AND DISCUSSION

Radionuclide daughters produced by Cf-252 decay consist of hundreds of radioisotopes. Several of them which has significant radioactivity are listed in Table 1. One important radioisotope produced by Cf-252 decay is curium-248 (Cm-248) which is a very beneficial radioisotope in research purposes due to its long half-life (3.40 × 105 years). This radioisotope can be extracted after several years decay of Cf-252. From Table 1, it can be seen that 0.49µCi of Cm-248 can be extracted after 6 months decay of Cf-252 and 1.6 mCi of Cm-248 can be extracted after 2 years decay of Cf-252. This amount of radioactivity is a bit significant to be used in research purposes.

Figure 1 shows the neutron flux produced from Cf-252 decay in the several time intervals between 0.1 year and 10 years. These neutrons are originated from spontaneous fission reactions of Cf-252 as well as from (α,n) reactions of other radionuclide daughters of Cf -252. From this graph it can be seen that one milligram Cf-252 produces a very high number of neutrons which is on the order of 109 neutrons per second. Moreover, even after several years of decay, the neutron fluxes from Cf-252 decay are maintained at high intensity. This makes Cf-252 as a reliable neutron sources for many applications. For the application of nuclear reactors start-up, a neutron source on the order of 109 neutrons per second is suitable for large reactors of about 1000 MWe.

Table 1. Several radioisotopes produced from Cf-252 decay

Decay Time (years)	Radioactivity (curie)				
	U-240	Pu-240	Np-240m	Cm-248	Cf-252
0.1	4.06E-17	1.41E-22	4.06E-17	1.05E-07	5.24E-01
0.5	4.06E-17	1.74E-20	9.81E-16	4.99E-07	4.72E-01
1	3.76E-16	1.35E-19	3.76E-15	9.37E-07	4.12E-01
2	1.38E-16	1.02E-18	1.39E-14	1.66E-06	3.18E-01
5	6.91E-15	1.34E-17	6.91E-14	2.97E-06	1.45E-01
8	1.45E-15	4.72E-17	1.45E-13	3.56E-06	6.58E-02
10	2.01E-14	8.39E-17	2.02E-13	3.76E-06	3.89E-02

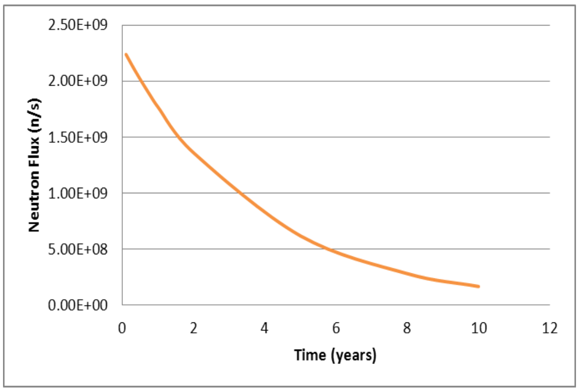

Fig. 1. Neutron flux produced from californium -252 decay in 10 years

Figure 2 shows the results for alpha radioactivity of Cf-252 decay. Alpha radioactivity after the first three months Cf-252 decay is 0.5 Ci. It is a high radioactivity since Cf-252 is a highly alpha emitter (96.91%) with short half-life (2.6 years). From this result it can be seen that the alpha radioactivity decreases slowly and its radioactivity is still in a significant amount even after Cf-252 decays for 10 years.

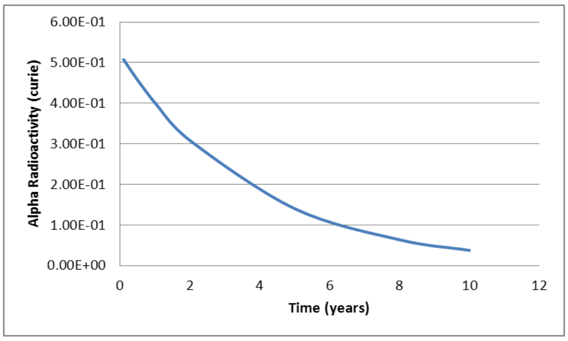

Fig. 2. Alpha particle radioactivity in 10-years decay of Cf-252

CONCLUSIONS

The calculation of radionuclide content by using ORIGEN2.1 computer code has been used in many purposes particularly in the aspect of nuclear fuel cycle management. The capability of ORIGEN2.1 to calculate not only radionuclide composition but also some important characteristics of nuclear materials is the reason why it is used widely in the world and known as a reliable code. In case of californium-252 radionuclide, one of radionuclide composition resulting from 10-years decay simulated by ORIGEN2.1 will consist of curium-248 as beneficial radioisotope for research purpose. The calculation of neutron fluxes also gives valuable information that californium-252 produces a highly neutron intensity, which makes californium-252 as a potential neutron source for many applications.

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