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Application of Origen2.1 in the decay photon spectrum calculation of spallation products*

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Abstract: Origen2.1 is a widely used computer code for calculating the burnup, decay, and processing of radioactive materials. However, the nuclide library of Origen2.1 is used for existing reactors like pressurized water reactors. To calculate the photon spectrum released by the decay of spallation products, we have made specific libraries for the ADS tungsten spallation target, based on the results given by the FLUKA Monte Carlo code. All the data used to make the Origen2.1 libraries are obtained from Nuclear structure & decay Data (NuDat2.6). The accumulated activity of spallation products and the contribution of nuclides to photon emission are given in this paper.

Keywords: Origen2.1, FLUKA, decay and photon libraries, spallation products, decay photon spectrum, NuDat2.6

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1 Introduction

Accelerator Driven subcritical Systems (ADS), consisting of proton accelerator, spallation target and subcritical reactor, are a widely proposed way to transmute high-level radioactive waste (HLW) produced by nuclear reactors [1]. To provide external neutrons for the subcritical reactor, the spallation target needs to be irradiated by high energy protons for a long time, which will lead to the accumulation of a large amount of radioactive products. So shielding after the decommissioning of the spallation products is necessary. Shielding against gamma rays is the most important due to their strong penetrability. Here we use Origen2.1 to calculate the decay photon spectrum of spallation products to provide some reference data for the shielding of decommissioned spallation targets.

FLUKA is a widely used Monte-Carlo code for simulating the interaction and transport of hadrons, heavy ions and electromagnetic particles from thermal neutron energies (100 eV) to cosmic ray energies (TeV) in any kind of material [2]. In ADS studies, FLUKA is used to calculate the spallation neutron spectrum, accumulation of residual nuclei and design of the proton accelerator. It provides a card called RESNUCLE to calculate the spallation products. For low energy (<5 GeV) projectiles like neutrons, protons and pions, a hadronic model of FLUKA, called PEANUT, is used to simulate the pre-equilibrium stage. For the de-excitation stage, a combination of models like self evaporation (or GEM), fission, multifragmentation and Fermi breakup are used to model the spallation reactions [3, 4]. Here we use FLUKA to calculate the accumulated activity of spallation products and provide it to Origen2.1 to calculate the decay photon spectrum.

Origen2.1 is a point-depletion and radioactive-decay computer code to simulate nuclear fuel cycles and calculate the nuclide compositions and characteristics of the materials used [5]. It was developed by Oak Ridge National Laboratory (ORNL) and has been used widely around the world since the 1970s. It can be used to simulate fuel cycle flow sheets and burnup of specific nuclides with the use of various corresponding databases [6].

Since Origen2.1 is used for existing reactors, it is not appropriate to apply Origen2.1 to calculate the decay photon spectrum of spallation products directly. So we have made specific libraries for a tungsten spallation target, containing the information of decay and photon production rates.

2 Calculation method & model

The calculation flow can be described as follows. Firstly, we build a calculation model of spallation tar-

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get by FLUKA, and calculate the accumulated activity of spallation products. Secondly, we make the decay and photon libraries of relative isotopes, based on the results given by FLUKA. Finally, we put the accumulated mass (in unit of grams) yield and the libraries together to the Origen2.1 input and get the 18 energy group photon spectrum. Figure 1 gives an intuitive view of the calculation flow.

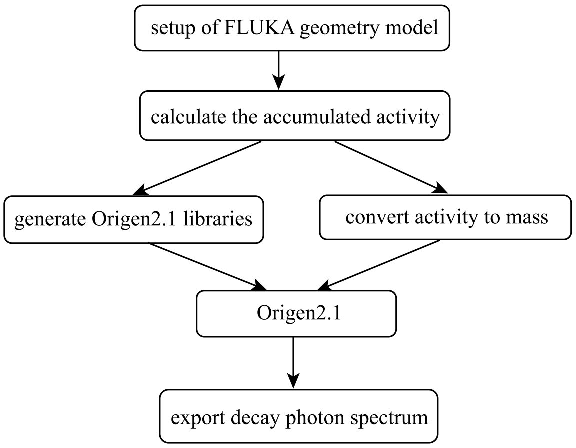

Fig. 1. Calculation flow of decay photon spectrum

2.1 Calculation of spallation products

To generate the Origen2.1 libraries, we need to know the details of the spallation products. The geometry structure of spallation target is shown in Fig. 2. The target is a cylinder with a proton beam pipe inside it. Protons generated from a linear accelerator will bombard the spallation material at a depth of 80cm. The thickness of the beam and target tubes are 0.5 cm and 1 cm respectively. The spallation target consists of tungsten, ion and nickel and the pipe wall material is T91.

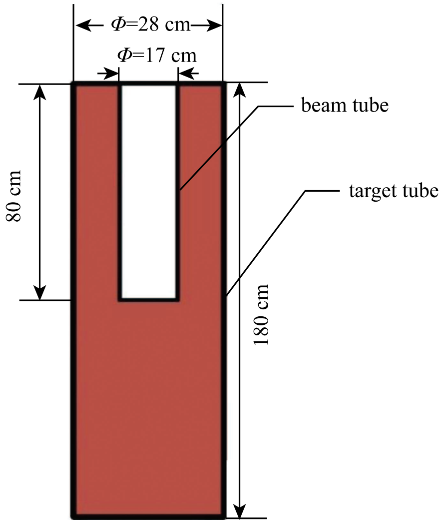

Fig. 2. Geometry structure of spallation target.

Here, we calculate the accumulated activities of the radioactive products combining the RADDECAY, RESUCLE, DCYTIMES, DCYCSCORE, and IRRPROFI card of FLUKA. The new evaporation model is activated to ensure a better quality of the results. Products scored here include spallation products (all interactions except those induced by neutrons below the threshold for multigroup treatment) and low-energy neutron products. The main parameters of the calculation model are given in Table 1. The preliminary number of particles here is set to 1×10^7.

Table 1. Calculation parameters.

parameter	material/Value
energy of the protons	250 MeV
current of the accelerator	10 mA
proton beam distribution	uniform
radius of beam spot	5 cm
spallation target form	granular
spallation material	Tungsten93-Iron4.5-Nickel2.5 (mass fraction %)
equivalent density	9.42 g/cm^3
irradiation time	5 a

Since Origen2.1 input requires the mass yield of nuclides, the accumulated activity can be easily transferred to mass yield by the equation below:

$$Y_i=\frac{A_iM_i}{\lambda_iN_\text{A}}. \tag{1}$$

where Y_i is the mass yield of nuclide i, A_i is the activity given by FLUKA, M_i is mass number, λ_i is the radioactive decay constant and N_A is Avogadro's constant.

2.2 Generation of Origen2.1 libraries

Origen2.1 external libraries consist of decay library, photon release library and cross-section library. The cross-section library is used to calculate the transmutation of wastes. Since we neglect the transmutation of products by neutrons, it is not necessary when calculating the decay photon spectrum. Information like half-life, decay branching ratio, average recovery energy and inhalation limits is included in the decay library. At present, we added half-life, decay branching ratio to the decay library. Other values here are set to zero. For convenient calculation, the photon energies in Origen2.1 are divided into 18 groups. Table 2 gives the concrete energy group structures of the photon library.

To generate the decay and photon release libraries, we get data from Nuclear Structure & decay Data (NuDat2.6), and reprocess these data into the corresponding format requested by Origen2.1. Since the photon release rate given by NuDat2.6 is discrete in energy, we count the release rate falling in each region listed above, and obtain the photon library.

Table 2. Origen2.1 photon library structures.

group	lower boundary/MeV	upper boundary/MeV	average energy/MeV
1	0.0000E-02	2.0000E-02	1.0000E-02
2	2.0000E-02	3.0000E-02	2.5000E-02
3	3.0000E-02	4.0000E-02	3.7500E-02
4	4.0000E-02	7.0000E-02	5.7500E-02
5	7.0000E-02	1.0000E-01	8.5000E-02
6	1.0000E-01	1.5000E-01	1.2500E-01
7	1.5000E-01	3.0000E-01	2.2500E-01
8	3.0000E-01	4.5000E-01	3.7500E-01
9	4.5000E-01	7.0000E-01	5.7500E-01
10	7.0000E-01	1.0000E 00	8.5000E-01
11	1.0000E 00	1.5000E 00	1.2500E 00
12	1.5000E 00	2.0000E 00	1.7500E 00
13	2.0000E 00	2.5000E 00	2.2500E 00
14	2.5000E 00	3.0000E 00	2.7500E 00
15	3.0000E 00	4.0000E 00	3.5000E 00
16	4.0000E 00	6.0000E 00	5.0000E 00
17	6.0000E 00	8.0000E 00	7.0000E 00
18	8.0000E 00	1.0000E 01	9.5000E 00

2.3 Calculation of decay photon spectrum

When calculating the decay photon spectrum, the accumulated nuclei of specific nuclides is necessary. In Origen2.1, how the amount of nuclide N_i changes as a function of time is described by a nonhomogeneous first-order ordinary differential equation as follows [6]:

$$\frac{\mathrm{d}N_i}{\mathrm{d}t}=\sum_{j=1}^{N}l_{ij}\lambda_jN_j+\phi\sum_{k=1}^{N}f_{ik}\sigma_kN_k -(\lambda_i+\phi\sigma_i+r_i)N_i+F_i, i=1\cdots N, \tag{2}$$

where

N_i= atom density of nuclide i

N= number of nuclides

l_{ij}= fraction of radioactive disintegration by other nuclides, which leads to formation of nuclide i;

λ_i =radioactive decay constant;

Φ= position and energy-averaged neutron flux, in calculation it is set to 0;

f_{ik}= fraction of neutron absorption by other nuclides, which leads to formation of nuclide 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 nuclides i;

By solving these N equations, Origen2.1 gives the amounts of each nuclide at the end of each time step set in the DEC card. The photon release rate by nuclide i thus can be calculated from the equation:

$$P_i=g_i\lambda_iN_i. \tag{3}$$

where P_i is the photon release rate of group i, g_i is the photon released per decay, which is given in Origen2.1 photon library, and λ_i and $N_i$are described above.

After getting the spallation yield and Origen2.1 libraries, the 18 energy group photon release data can be easily obtained using the DEC card.

3 Results and discussion

3.1 Accumulated activity of spallation products

Based on the geometry and calculation parameters mentioned above, the accumulated activity of spallation products can be calculated by FLUKA. The spallation material consists of natural tungsten and iron/nickel. Table 3 gives the main radioactive isotopes produced in the target region. Here, the nuclide ID is defined as $1000A+Z$, where A is the mass number and Z is the atomic number. The products scored here include all spallation products and low-energy neutron products. The activity of tungsten products is greater than iron/nickel by an order of one magnitude.

We can see that the main radiation in the products is induced by the isotopes of spallation materials as shown in Fig.3. There are three peaks in the distribution of radioactive products, which are induced by light evaporation fragments, iron/nickel and tungsten. The highest activity appears at atomic number 74, and reaches nearly 5×10^{15} Bq. The peak caused by iron/nickel is approximately 5×10^{14} Bq.

Table 3. Main radioactive products of spallation materials.

spallation material	main products (nuclide ID)	activity/Bq	error (%)1)	half-life/d
W^{nat} (93%)2)	74185	4.97E+15	0.1338	75.10
	74181	4.09E+15	0.1176	121.20
	74187	2.81E+15	6.43E-02	0.99
	74179	2.52E+15	0.1052	0.03
	73179	2.47E+15	0.1185	664.30
Fe^{nat}(4.5%)/Ni^{nat}(2.5%)	26055	4.40E+14	0.3758	999.01
	27058	3.63E+14	0.2497	70.86
	27057	2.75E+14	0.4293	271.74
	25054	2.33E+14	0.3785	312.03
	24051	1.55E+14	0.5208	27.70

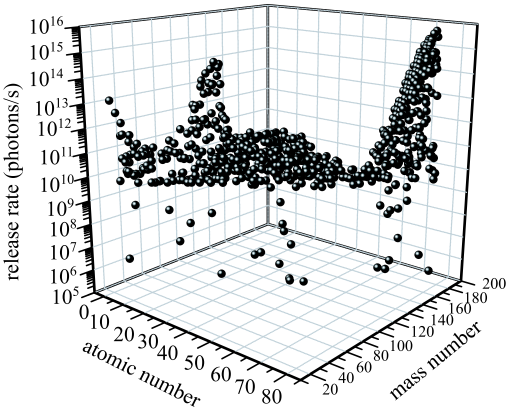

Fig. 3. Distribution of radioactive products.

3.2 Decay photon spectrum

The decay library we have made includes about 370 kinds of spallation products and the photon library contains about 260 kinds of nuclides which have activity higher than 1×10^{12} Bq. After generating the Origen2.1 libraries, we calculated the decay photon release rate at different cooling times. The results are shown in Table 4. The main photon emitters are isotopes of W, Ta, Lu, Re, Yb and Hf, most of which have short half-lives and thus contribute little to photon emission after a comparatively long cooling time. For nuclides like ^{44}Ti, ^{90}Y, ^{157}Tb, ^{94}Nb, and their daughters, they contribute more than 90% to photon emission after a cooling time of 100 a, and the total photon release rate decreases about 5 orders of magnitude. The total gamma power is 3.71×10^3 W, which is relatively low and thus is negligible.

The decay photon spectrum calculated by Origen2.1 at different cooling times is shown in Fig. 4. Most photons released by decay of spallation products are in range of 0—1 MeV. The high energy group photon release rate is negligible. After cooling times of 1 year and 100 years, the photon release rate will decrease about 10 and 10^6 times respectively.

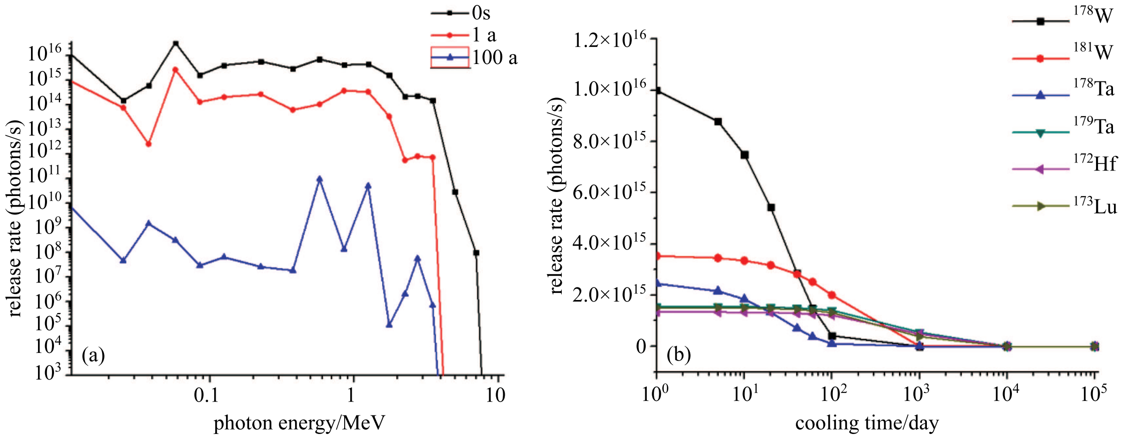

Fig. 4. (a) Decay photon spectra at different cooling times (b) Change of total photon release rate of main photon emitters with respect to cooling time.

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1) Statistical error of activity in percentage given by FLUKA, that is $\sigma/N^{1/2}$, where σ stands for standard deviation, N stands for number of events.

2) Mass fraction, superscript “nat” stands for natural

Table 4. 18 energy group decay photon spectrum at different cooling time.

$E_{\rm mean}$/MeV\cooling time	0 s	1 d	100 d	1 a	100 a
1.00E-02	1.75E+16	1.00E+16	2.29E+15	1.17E+15	1.17E+10
2.50E-02	1.51E+14	1.24E+14	1.02E+14	7.62E+13	4.56E+07
3.75E-02	6.15E+14	1.14E+14	5.41E+12	2.58E+12	1.46E+09
5.75E-02	3.37E+16	1.92E+16	5.21E+15	2.70E+15	3.12E+08
8.50E-02	1.64E+15	6.95E+14	2.05E+14	1.32E+14	2.91E+07
1.25E-01	4.09E+15	1.57E+15	4.34E+14	2.07E+14	6.39E+07
2.25E-01	5.86E+15	1.91E+15	6.96E+14	2.72E+14	2.58E+07
3.75E-01	3.02E+15	8.43E+14	1.84E+14	6.27E+13	1.85E+07
5.75E-01	7.16E+15	2.21E+15	3.40E+14	1.06E+14	9.48E+10
8.50E-01	4.16E+15	1.91E+15	7.33E+14	3.76E+14	1.34E+08
1.25E+00	4.57E+15	1.72E+15	5.80E+14	3.41E+14	5.07E+10
1.75E+00	1.62E+15	3.48E+14	5.29E+13	3.40E+13	1.09E+05
2.25E+00	2.24E+14	9.58E+13	6.09E+12	5.62E+11	2.06E+06
2.75E+00	2.36E+14	9.02E+13	8.84E+12	8.17E+11	5.62E+07
3.50E+00	1.54E+14	3.27E+13	7.90E+12	7.30E+11	7.02E+05
5.00E+00	2.88E+10	1.48E+09	6.85E+03	1.72E-07	1.72E-07
7.00E+00	9.77E+07	1.11E-08	1.11E-08	1.11E-08	1.11E-08
9.50E+00	7.05E-10	7.05E-10	7.05E-10	7.05E-10	7.05E-10
TOTAL	8.48E+16	4.09E+16	1.09E+16	5.48E+15	1.59E+11
γ power (W)	3.71E+03	1.35E+03	3.73E+02	1.87E+02	1.90E-02

Table 5. Photon release rate of main photon emitters in energy group 1.

photon emitter\cooling time	0 s	1 d	100 d	1 a	100 a
^{178}W	4.69E+15	4.54E+15	1.89E+14	3.81E+10	0.00E+00
^{73}As	1.78E+15	0.00E+00	0.00E+00	0.00E+00	0.00E+00
^{177}W	1.02E+15	5.55E+11	0.00E+00	0.00E+00	0.00E+00
^{181}W	7.74E+14	7.70E+14	4.37E+14	9.59E+13	0.00E+00
^{179}W	7.30E+14	2.03E+03	0.00E+00	0.00E+00	0.00E+00
^{176}Ta	6.44E+14	1.13E+14	0.00E+00	0.00E+00	0.00E+00
^{175}Ta	5.25E+14	1.13E+14	0.00E+00	0.00E+00	0.00E+00
^{174}Lu	5.21E+14	4.22E+14	4.19E+05	0.00E+00	0.00E+00
^{178}Ta	5.19E+14	4.70E+14	1.96E+13	3.94E+09	0.00E+00
^{177}Ta	5.15E+14	3.97E+14	9.16E+01	0.00E+00	0.00E+00
^{179}Ta	4.99E+14	4.98E+14	4.49E+14	3.41E+14	1.44E-02
^{172}Hf	4.07E+14	4.06E+14	3.67E+14	2.81E+14	3.25E-02
^{174}Ta	3.85E+14	3.11E+08	0.00E+00	0.00E+00	0.00E+00
^{173}Ta	3.57E+14	1.84E+12	0.00E+00	0.00E+00	0.00E+00
^{171}Lu	3.20E+14	3.08E+14	7.57E+10	1.55E+01	0.00E+00
^{181}Re	3.18E+14	1.38E+14	0.00E+00	0.00E+00	0.00E+00
^{173}Lu	3.07E+14	3.07E+14	2.68E+14	1.85E+14	3.27E-08
^{180}Re	2.54E+14	3.15E-10	0.00E+00	0.00E+00	0.00E+00
^{179}Re	2.53E+14	0.00E+00	0.00E+00	0.00E+00	0.00E+00
^{44}Sc	7.33E+10	2.41E+09	1.31E+09	1.30E+09	4.14E+08
^{44}Ti	3.01E+10	3.01E+10	3.00E+10	2.97E+10	9.48E+09
^{90}Y	2.49E+10	1.96E+10	1.79E+09	1.76E+09	1.67E+08
^{157}Tb	2.24E+09	2.24E+09	2.23E+09	2.23E+09	1.41E+09

4 Conclusions

In our work, specific Origen2.1 libraries for a tungsten spallation target have been made for further relative calculation. The accumulated activity of products induced by a 250 MeV, 10 mA proton beam is calculated by the Monte-Carlo code FLUKA, and the distribution of activity with atomic and mass numbers are given above. The main decay photon release rate calculated by Origen2.1 is at a magnitude of 10^{15}, and the energy is concentrated in the low energy region, that is 0—1 MeV (accounting for 91.86% of the total).

The main photon emitters in photon group 1 is given in Table 5. For all 18 groups of photons, the results show that the main photon emitters like ^{177}W, ^{177}Ta and ^{73}As are of relatively short half-life, and will decrease rapidly, which can be seen in Fig. 4(b) intuitively. After a long cooling time, the main emitters will be ^{44}Ti, ^{90}Y, ^{157}Tb, ^{94}Nb, and their daughters.

After a cooling time of 100 years, the total photon release rate will decrease from 8.48×10^{16} to 1.59×10^{11} photons/s. Thus, there is no need to worry about long term disposal for the spallation target from the aspect of gamma ray shielding.

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