Monte Carlo modeling of spallation targets containing uranium and americium

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Abstract

Neutron production and transport in spallation targets made of uranium and americium are studied with a Geant4-based code MCADS (Monte Carlo model for Accelerator Driven Systems). A good agreement of MCADS results with experimental data on neutron- and proton-induced reactions on $^{241}$Am and $^{243}$Am nuclei allows to use this model for simulations with extended Am targets. It was demonstrated that MCADS model can be used for calculating the values of critical mass for $^{233}$U, $^{235}$U, $^{237}$Np, $^{239}$Pu and $^{241}$Am. Several geometry options and material compositions (U, U+Am, Am, Am$_2$O$_3$) are considered for spallation targets to be used in Accelerator Driven Systems. All considered options operate as deep subcritical targets having neutron multiplication factor of $k \sim 0.5$. It is found that more than 4 kg of Am can be burned in one spallation target during the first year of operation.

Keywords: spallation reactions, minor actinides, neutron sources, Accelerator Driven Systems, radioactive waste

1. Introduction

Many neutrons can be produced in spallation nuclear reactions \cite{1,2} induced by energetic protons in collisions with heavy target nuclei like W, Ta, Bi and Pb due to their enhanced neutron content with respect to lighter nuclei. This method to create an intense flux of neutrons is known for decades and it is already employed in several existing \cite{3,4} spallation neutron sources and will be used in the facilities to be constructed, e.g., in the ESS project \cite{5}. Such facilities are dedicated to neutron imaging and scattering experiments \cite{6}. Accelerator Driven Systems (ADS) aimed at energy production in subcritical assemblies of fissionable materials or burning nuclear waste \cite{7,8} also use an intense proton beam to produce neutrons in spallation targets. The design of a spallation target is a challenging part of such projects in view of high energy deposited by the proton beam and secondary particles and the radiation damage of the target material. The performance of a target irradiated by a megawatt-power proton beam was the subject of a dedicated experiment \cite{9}.

Heavy materials like W, Ta, Bi and Pb are commonly used in the design of spallation targets. Although the fission of such nuclei can, in principle, be induced by energetic protons \cite{10}, its role in neutron production is negligible. However, an alternative approach can be also considered to involve fissionable, $^{232}$Th, $^{238}$U \cite{11}, or even fissile, $^{235}$U, $^{242}$mAm \cite{12}, materials in the design of a spallation target. The difference between these two groups of materials consists in the capability of fissile material to sustain a nuclear chain reaction once a critical mass of this material is accumulated. Such materials can be either directly irradiated by a proton beam, or used as a blanket surrounding a non-fissionable material impacted by protons. In both cases neutron production is boosted due to additional fission neutrons. As recently demonstrated by our calculations \cite{13}, the number of neutrons produced per beam proton is about 3 times higher in a uranium target compared to one made of tung...
sten, while the energy deposition calculated per produced neutron remains comparable in both targets. Therefore, a less powerful beam is needed to achieve the same neutron flux in the uranium target as in the tungsten target, and the total energy deposition in both targets \[23\] remain comparable. Thermal energy released in fission reactions can be converted to electricity and then support, at least in part, the operation of the accelerator.

Apart from the need to build intense neutron sources, using fissile materials in spallation targets opens the possibility to transmute them in fission reactions induced by primary protons and secondary nucleons. Indeed, in addition to unused uranium, each 1000 kg of spent nuclear fuel discharged from a light-water reactor typically contain several kilograms of fissile transuranium elements like plutonium and Minor Actinides (MA): neptunium, americium and curium \[14\]. Up to 99.9% of plutonium can be extracted and then further used in nuclear reactors \[15\]. However, other radioactive elements, MA and long-lived fission products, are still very hazardous due to their high radiotoxicity, and their release to environment has to be avoided. There are plans to confine them in very robust vitrified blocks stored in deep geological repositories. Alternatively, MA contained in spent nuclear fuel can be separated and recycled in a dedicated facility operating with fast neutrons (as thermal neutrons are not efficient). As demonstrated by many dedicated studies, see e.g. \[14\], the extracted MA can be efficiently transmuted into short-lived or stable fission products in fast reactors or in accelerator-driven reactor cores.

Certainly, more theoretical and experimental studies are needed to design an intense fast-neutron source or a spallation target containing fissionable or fissile materials. For many years experimental studies of transmutation of long-lived radiotoxic nuclides have been carried out at the Joint Institute for Nuclear Research in Dubna, Russia, in the framework of an international collaboration \[16\]. In particular, \[237\]Np and \[241\]Am were transmuted into short-lived or stable nuclides by neutrons produced by protons in a thick lead target. Within the project called “Energy plus Transmutation” beams of protons and neutrons were used, and the flux of fast neutrons was amplified by a massive uranium sleeve surrounding a non-fissile target \[13\] \[17\] \[18\].

Detailed theoretical modeling of ADS prototypes should precede their construction and operation. Therefore, a reliable computational tool based on modern software is necessary to foster studies in the field of the accelerator-driven transmutation. A number of Monte Carlo codes have been used to simulate neutron production and transport in spallation targets of ADS: PHITS \[19\], SHIELD \[20\], MCNPX \[21\] and others. However, to the best of our knowledge, spallation targets containing Am were not studied with these codes so far. In the present work we further develop our Geant4-based code MCADS (Monte Carlo model for Accelerator Driven Systems) \[13\] \[22\] in order to apply it for fissile spallation targets containing U and Am. Modeling spallation targets containing americium is motivated by the following two reasons \[14\]. First, americium is the most abundant MA in spent nuclear fuel and its transmutation into relatively short-lived fission products can reduce the radiotoxicity of radioactive waste by an order of magnitude. Second, the operation of fast reactors with a high content of MA causes certain safety concerns. Alternatively, a subcritical system driven by an accelerator could be a promising option to burn americium extracted from spent nuclear fuel.

2. Modeling of americium transmutation by slow and energetic nucleons

As demonstrated in our previous works \[13\] \[22\], all physics processes relevant to neutron generation and transport in conventional non-fissile and also in fissionable uranium targets can be successfully simulated with the Geant4 toolkit \[23\] \[24\] \[25\]. In particular, these processes include spallation and fission reactions induced by primary protons and secondary nucleons. Usually specific Geant4 simulations are performed with a set of physical models known as a Physics List.

All present calculations were performed with Geant4 of version 9.4 with patch 01 as in our previous works \[13,22\]. In this version of the toolkit the following models are available for simulating p-nucleus interactions: Bertini Cascade, Binary Cascade and Intra-Nuclear Cascade Liège coupled with the fission-evaporation model ABLA. These models are included in the QGSP_BERT_HP, QGSP_BIC_HP and QGSP_INCL_ABLA Physics Lists, respectively. The prefix QGSP indicates that quark-gluon string model is used for high-energy interactions. All three Physics Lists employ High Precision (HP) model for neutron interactions below 20 MeV which use evaluated nuclear data libraries described below. The ionization energy loss
of charged particles was simulated with Standard Electromagnetic Physics package of Geant4. The physics models used in Geant4 are described in detail in Geant4 Physics Manual [26].

In Ref. [13] we have evaluated the performance of the above-mentioned physics models for tungsten and uranium targets irradiated by protons. Fission cross sections and multiplicities of neutrons produced in thin uranium targets by protons with energies of 27, 63 and 1000 MeV were calculated and compared with experimental data [27, 28]. It was demonstrated, that the INCL\_ABLA [29, 30] better describes the data as compared with other models. In particular, only INCL\_ABLA predicts the fission cross section and the neutron multiplicity for 1000 MeV protons very close to data, within the uncertainty of the measurements. However, one can note that all the considered cascade models become less accurate for proton energies below 100 MeV [13].

The average numbers of neutrons produced in extended tungsten and uranium targets irradiated by 400-1500 MeV protons were also calculated and compared with experimental data, see Ref. [13]. As shown, also in this case the combination of INCL\_ABLA and Neutron\_HP models provides the most accurate results, which differ by less than 10% from the experimental data. The Bertini Cascade model mostly overestimates, while the Binary Cascade model underestimates the neutron yields. Therefore, we conclude that the QGSP\_INCL\_ABLA\_HP Physics List is the best choice among other options for simulating nuclear reactions in uranium and tungsten targets.

In order to perform simulations with materials containing americium several extensions of the Geant4 toolkit have been introduced in [31]. This made possible the simulations of proton- and neutron-induced nuclear reactions and elastic scattering of nucleons on Am and other transuranium nuclei. In our recent publication [32] the (p,f), (n,f) and (n,γ) cross sections as well as mass distributions of fission fragments, average number of neutrons per fission event and secondary neutron spectra were calculated with MCADS for 241\textsuperscript{Am} and 243\textsuperscript{Am}, and good agreement with experimental data was obtained. This justifies using MCADS to simulate extended targets containing 241\textsuperscript{Am} and 243\textsuperscript{Am}.

The physics of transmutation of 241\textsuperscript{Am} and 243\textsuperscript{Am} nuclei by neutron irradiation can be well understood from Fig. 1. Depending on the neutron energy both nuclei can either undergo fission or be transformed via (n,γ) reaction into A+1 isotopes 242\textsuperscript{Am} and 244\textsuperscript{Am}. After β⁻-decay with half-life times 16 h and 10 h these nuclei change finally into long-lived 242\textsuperscript{Cm} and 244\textsuperscript{Cm}, respectively. A sharp rise of the fission cross section at incident neutron energy of ~ 0.6 MeV leads to the dominance of fission of 241\textsuperscript{Am} and 243\textsuperscript{Am} over the neutron capture above 1 MeV. Therefore, fast neutrons produced in primary spallation reactions and subsequent neutron-induced fission reactions can be used to burn 241\textsuperscript{Am} and 243\textsuperscript{Am} very efficiently.

Figure 1: Radiative neutron capture cross section (n,γ), shown in red, and neutron-induced fission cross section (n,f), shown in blue, for 241\textsuperscript{Am} (top) and 243\textsuperscript{Am} (bottom) nuclei. MCADS results are represented by solid lines, cross sections from ENDF/B-VII.1 evaluated nuclear data library – by dashed lines. Measured cross sections, (n,γ) for 241\textsuperscript{Am} and 243\textsuperscript{Am} from Refs. [33, 34] are shown by triangles, and (n,f) cross section from Refs. [35, 36] – by squares.

The radiative neutron capture (n,γ) and fission (n,f) cross sections calculated by MCADS by means...
of the Monte Carlo modeling of neutron interactions with a thin layer of $^{241}$Am or $^{243}$Am are plotted in Fig. 1 together with the corresponding experimental data [33, 34, 35, 36]. Nuclear reactions induced by neutrons with energy below 20 MeV are simulated by MCADS on the basis of the evaluated nuclear data library JENDL-4.0 [37] converted into a format readable by Geant4 [38]. It was found that the Geant4-compatible nuclear data files based on JENDL-4.0 provide the most accurate description of the energy spectra of secondary neutrons with respect to other nuclear data libraries. The MCADS results below 20 MeV can be compared to the cross sections extracted from ENDF/B-VII.1 [39], which are also shown in Fig. 1. As seen from this figure, a very good agreement is obtained between MCADS, experimental data and ENDF/B-VII.1 data.

3. MCADS calculations of neutron multiplication in fissionable materials

The key issue in designing a spallation target containing fissionable materials is the calculation of the neutron multiplication factor to ensure that the target operates in a safe subcritical regime. Neutron multiplication factor $k$ is calculated with MCADS as the ratio between the numbers of neutrons in the present and previous generations of neutrons averaged over many simulated events. The obvious condition is to keep $k < 1$, i.e. strictly in the subcritical mode. The number of neutrons in the target is determined by the balance between their production, absorption and leakage through the target surface.

In order to validate the MCADS model with respect to generation of fission neutrons and their absorption in $(n,\gamma)$ and $(n,f)$ processes, we have performed simulations of neutron multiplication in bare (unreflected) spheres made of several fissile materials listed in Table 1. The radius of each sphere made of specific material was gradually increased until $k$ asymptotically exceeded 1, and the mass of each sphere was determined as the critical mass for the given material, see Table 1. The critical mass data published by the European Nuclear Society [40] and Monte Carlo simulation results obtained with JENDL-3.2 library for $^{237}$Np and $^{241}$Am in Refs. [11] and [42] are also presented in Table 1. The results of calculations with MCADS for $^{233,235}$U, $^{237}$Np and $^{239}$Pu agree within 2–8% with the published data [40], but diverge by ~16% for $^{241}$Am. We attribute this deviations to uncertainties of the nuclear data for $^{241}$Am.

Following the validation of MCADS results for the critical mass of the $^{243}$Am sphere without external irradiation, we investigated the criticality issues for a cylindrical spallation target made of pure $^{241}$Am and irradiated by a proton beam. The length of the target was fixed at 150 mm to ensure that all beam protons are stopped in the target material, while the target radii were varied from 40 mm to 110 mm. It was assumed that the target was irradiated by a 600 MeV proton beam with the transverse beam profile of 20 mm FWHM. In Fig. 2 we show the time dependence of the average number of neutrons inside the targets with the target radii of 40, 60, 80, 100, 106 and 110 mm. One can see that the average number of neutrons in the targets with radii 40–100 mm decreases at late time because less neutrons are produced inside the target volume than escape it or lost in nuclear interactions. The number of neutrons saturates in the target of 106 mm radius (with the weight of 72.4 kg) just 30 ns after the impact of a beam proton. This case is very close to the critical regime with $k = 0.999$. Even a smaller fraction of neutrons escape a thicker target of 110 mm radius, and this target becomes supercritical ($k = 1.013$). The corresponding neutron multiplication factors are listed in the legend of Fig. 2.

From this criticality study we can conclude that thin cylindrical $^{241}$Am targets with typical radii of ~50 mm and length of 150 mm irradiated by 600 MeV protons will operate in a deep subcritical regime with $k \sim 0.7$. This suggests that an equivalent amount of $^{241}$Am can be used to build a fissile spallation target. All geometrical configura-
Table 1: Calculated critical masses (kilograms) of bare spheres made of $^{233,235}\text{U}$, $^{237}\text{Np}$, $^{239}\text{Pu}$ and $^{241}\text{Am}$. Data from European Nuclear Society [40] and Monte Carlo modeling results by Polina [41] and MCNP [42] codes both based on JENDL-3.2 library are given for comparison.

| Material | MCADS results | data [40] | calculations [41] | calculations [42] |
|----------|----------------|-----------|-------------------|-------------------|
| $^{233}\text{U}$ | 16.1 | 15.8 | |
| $^{235}\text{U}$ | 48.4 | 46.7 | |
| $^{237}\text{Np}$ | 62.4 | 63.6 | 75.0 |
| $^{239}\text{Pu}$ | 10.8 | 10.0 | |
| $^{241}\text{Am}$ | 66.7 | 57.6 | 71.8 | 73.7 |

Figure 2: Number of neutrons as a function of time in the cylindrical targets made of $^{241}\text{Am}$ with the length of 150 mm and various radii and irradiated by 600 MeV protons. The neutron number is normalized per beam particle. The steady-state behavior (horizontal line) corresponds to approaching critical regime with $k = 0.999$.

4. Comparison of targets containing uranium and americium

The operation of spallation targets containing uranium can be simulated with confidence, as the nuclear data for $^{nat}\text{U}$ and all its most abundant isotopes are reliable in all versions of nuclear data libraries. The simulations of pure uranium targets are very instructive for further comparison with U+Am and pure Am targets. Moreover, the neutrons from U fission can be used for the Am transmutation. Therefore, we have performed simulations of cylindrical targets made of $^{nat}\text{U}$, pure $^{241}\text{Am}$, a mixture of $^{241}\text{Am}$ and $^{243}\text{Am}$ (57% and 43%, respectively) and americium oxide $\text{Am}_2\text{O}_3$ with the same isotopic composition of Am. The choice of $\text{Am}_2\text{O}_3$ is motivated by the fact that americium is usually extracted from spent nuclear fuel in the form of americium oxide. Each target has the radius of 40 mm and length of 120 mm. All these targets have masses well below the critical mass given in Sec. 3. It was assumed that the targets were irradiated by the proton beam with the FWHM of 20 mm and the energy of 600 MeV.

The spatial distributions of neutron flux calculated with MCADS for the considered four targets are shown in Fig. 3. Although the results are given for the proton current of 10 mA, they can be easily rescaled to the actual beam current. The average neutron flux in the americium target (1.56 · 10$^{16}$ n/s/cm$^2$) is higher than in the uranium one (1.22 · 10$^{16}$ n/s/cm$^2$) due to a higher fission cross section for Am. Since the fission cross section on $^{241}\text{Am}$ is almost 3 times higher than on U, one could expect even a larger difference in favor of $^{241}\text{Am}$. However, other reactions, like $(n,2n)$, $(n,3n)$, $(n,4n)$, are much more probable on uranium nuclei. As the result, the difference in the average neutron flux between U and Am targets is reduced. As one can see from Fig. 3, the results for pure $^{241}\text{Am}$ and mixed $^{241}\text{Am}+^{243}\text{Am}$ targets are very similar.

Calculated spatial distributions of heat deposition inside the considered targets are presented in Fig. 4. As expected, in fissile spallation targets a significant energy is deposited due to fission reactions [22]. The difference between fission cross section on Am and U leads to significantly larger energy deposition in the americium targets (11.9–16.1 MW, depending on the isotope composition) compared to the uranium target (7.7 MW). Therefore, designing a cooling system for the Am target may cause a serious problem.

The values of the average neutron flux (1.32 ·
Figure 3: Distribution of the neutron flux inside cylindrical targets made of natU, 241Am, mixture of 241Am and 243Am and Am2O3, all of the same dimensions specified in Table 2. The targets are irradiated by a 10 mA 600 MeV proton beam.
Figure 4: Distribution of heat deposition inside cylindrical targets made of \( ^{235}U \), \( ^{241}Am \), mixture of \( ^{241}Am \) and \( ^{243}Am \) and \( Am_2O_3 \), all of the same dimensions specified in Table 2. The targets are irradiated by 600 MeV proton beam with current of 10 mA.
10^{16} \text{n/s/cm}^2\) and heat deposition (11.9 MW) calculated for a pure \textsuperscript{243}Am target are lower compared to a pure \textsuperscript{241}Am target. This is explained by a lower fission cross section of \textsuperscript{243}Am compared to \textsuperscript{241}Am, see Fig. 1. The corresponding values for the target made of the mixture of isotopes, \textsuperscript{241}Am+\textsuperscript{243}Am, are intermediate (1.44 \times 10^{16} \text{n/s/cm}^2 and 14 MW) with respect to the monoisotopic targets.

It is expected that an increased number of fission reactions in a spallation target makes possible to boost the transmutation rate of americium contained in the target. However, due to the additional energy released in fission events the heat deposition in the target rises too. As seen from Fig. 3 the energy deposition in U and \textsuperscript{241}Am targets in the hottest region located close to the target axis exceeds 100 kW/cm\textsuperscript{3}, which looks very problematic from technical point of view. Therefore, more sophisticated target systems with a reduced energy deposition per fissioned Am nucleus are needed.

By this reason we extended our calculations for two U targets containing \textsuperscript{241}Am as proposed in [13] and a pure \textsuperscript{242}Am target mentioned above. Two targets containing \textsuperscript{241}Am considered in [13] were schematically designed as following. In the first case \textsuperscript{241}Am was uniformly mixed with U with 10\% mass concentration and in the second case a cylindrical core \((V = 200 \text{ cm}^3)\) of \textsuperscript{241}Am is placed in the hottest region of the uranium target. In both cases the target has the radius of 10 cm and length of 20 cm. The heat deposition values per burned Am nucleus in these targets are several times higher than the corresponding values for the pure americium targets because of additional fission events of uranium nuclei which also produce neutrons.

The numbers of Am fission events per beam particle \(N_{fis}\) are listed in Table 2 for the six targets containing Am and also for the \textsuperscript{nat}U target taken as a reference case, where, accordingly, U fission events are counted. As seen from the table, depending on isotope composition 2 to 3 times more fission events per beam proton are estimated for the pure Am targets compared to the uranium target. Calculated energy deposition per fissioned Am nucleus, \(Q/N_{fis}\), is also given in Table 2. For the pure Am target this value (203 MeV) is by 30\% lower than for the uranium one (279 MeV). As expected, much more energy is deposited per Am fission event in two U+Am targets \((Q/N_{fis} = 901 \text{ MeV and 739 MeV})\), because only a part of fission events correspond to Am and most of them to U, according to their concentration in the targets.

Since \textsuperscript{Am}_2\text{O}_3 target contains less Am nuclei than the pure Am target, this leads to a lower number of fission events and results, correspondingly, in a lower neutron flux \((1.06 \times 10^{16} \text{n/s/cm}^2)\) and lower energy deposition (9.3 MW). The heat deposition per fission event \(Q/N_{fis} = 229 \text{ MeV} \) calculated for \textsuperscript{Am}_2\text{O}_3 target is close to the \(Q/N_{fis}\) value for the pure \textsuperscript{241}Am+\textsuperscript{243}Am target.

Finally, burning rates of Am calculated for 10 mA proton beam are presented in Table 2. It was assumed that the burning rates are proportional to the number of Am fission events \(N_{fis}\) in a corresponding target. As found, more than 0.5 kg of \textsuperscript{241}Am can be transmuted per month in the spallation target containing exclusively this isotope. Lower burning rates are estimated for other considered geometry and material options, mostly due to a lower Am content. The amount of americium transmuted during the first month of operation \(dm/dt(t = 0)\) can be used to calculate the amount of Am burned during the first year. In this estimation it is assumed that the amount of Am \(m(t)\) decreases exponentially, \(m(t) = m_0 \exp(-t/\tau)\), from its initial amount \(m_0\) with the characteristic time \(\tau = m_0/(dm/dt(t = 0))\). The corresponding results are listed in the last column of Table 2 which gives the minimum annual consumption of Am in the considered spallation targets. Indeed, a possibility to upload additional quantities of Am to the target \(\text{(e.g. on the regular monthly basis)}\) can be considered, thus substantially increasing the transmutation capability of the ADS system. One can conclude, that Am can be more efficiently burned in the spallation targets made of pure Am (more than 4 kg of Am per year) compared to mixed U+Am targets. However, the use of larger amounts of pure Am is restricted due to criticality issues.

As shown in our previous publication [13], in the spallation targets made of \textsuperscript{nat}U the neutron production is significantly enhanced with respect to the tungsten targets of the same size due to additional contribution of neutron-induced fission of uranium nuclei. In the present study we have found that even more neutrons are produced by fission reactions in targets made of pure \textsuperscript{241}Am and \textsuperscript{243}Am. This follows from Table 3 where the numbers of neutrons produced or absorbed in various nuclear reactions are given per beam particle for \textsuperscript{nat}U, \textsuperscript{241}Am and \textsuperscript{243}Am targets of the same size. This means that even small Am targets are highly efficient in incinerating Am in fission reactions. The number of neutrons which escape from the targets
Table 2: Initial amount of Am, number of fission reactions on Am per beam proton, $N_{fis}$, heat deposition per fissioned Am nucleus, $Q/N_{fis}$, the burning rate of Am at the beginning of operation and the amount of Am transmuted in the first year of operation for various target options.

| Material                  | Length (cm) | Radius (cm) | Initial Am mass (kg) | $N_{fis}$ | $Q/N_{fis}$ (MeV) | dm/dt (t = 0) (g/month) | Burned in first year (kg) |
|---------------------------|-------------|-------------|---------------------|-----------|------------------|------------------------|--------------------------|
| natU                      | 12          | 4           | 0                   | 2.74*     | 279*             | —                      | —                        |
| U+241Am (10%)             | 20          | 10          | 11.7                | 1.84      | 901              | 120                    | 1.4                      |
| U+241Am (core)            | 20          | 10          | 2.68                | 2.25      | 739              | 147                    | 1.6                      |
| Pure 241Am                | 12          | 4           | 8.24                | 7.91      | 203              | 514                    | 4.3                      |
| Pure 243Am                | 12          | 4           | 8.24                | 5.49      | 216              | 357                    | 3.4                      |
| 241Am+243Am               | 12          | 4           | 8.24                | 6.74      | 208              | 438                    | 3.9                      |
| $\text{Am}_2\text{O}_3$   | 12          | 4           | 6.46                | 4.07      | 229              | 265                    | 2.5                      |

* Number of fission reactions on U nuclei per beam proton. For all other target options $N_{fis}$ corresponds exclusively to Am fission events.

Table 3: Average contributions to neutron production and absorption from different reaction channels and the number of leaking neutrons for natU, 241Am and 243Am targets with the length of 12 cm and radius of 4 cm. All numbers are given per beam proton.

| Reaction | natU | 241Am | 243Am |
|----------|------|-------|-------|
| p + A    | 11.77| 9.01  | 9.34  |
| (n,2n)   | 0.44 | 0.04  | 0.11  |
| (n,3n)   | 0.15 | 0.01  | 0.02  |
| (n,4n)   | 0.09 | 0.06  | 0.06  |
| (n,>4n)  | 3.80 | 1.67  | 1.91  |
| (n,fission)* | 4.17 | 19.54 | 13.79 |
| produced by other particles | 0.15 | 0.26  | 0.17  |
| (n,γ)    | -0.45| -1.45 | -1.00 |
| leak      | 20.12| 29.14 | 24.40 |

* Only from neutron-induced fission below 20 MeV.

5. Spallation targets made of Am with U booster and Be reflector

Several advanced geometry options of the spallation target can be considered to increase the burning rate of Am by enhancing the neutron flux inside the target. Two such options are considered below. The first target option (b) consists of an 241Am cylinder covered by a 2 cm thick natU booster. The second option (c) is additionally covered by a 9Be reflector which is 10 cm thick. The core of the both target options consist of a 241Am cylinder (rod) with the length of 150 mm and radius of 20 mm, which is also used alone as a reference target option (a) for comparison. All three target options are schematically shown in Fig. 5 and their parameters are listed in Table 4.

Initially all the three targets contain four times less 241Am (2.58 kg) compared to the pure 241Am target considered in Sec. 4. However, the number of fission events per beam proton $N_{fis}$ in the target (c) is only twice as low as in the target of Sec. 4. This means that Am is burned more efficiently in the presence of the booster, or with both booster and reflector. At the same time $Q/N_{fis}$ calculated in the Am core for option (c) is comparable to the same parameter for the pure 241Am target consider-
Table 5: Initial amount of Am, number of fission events on Am per beam proton, $N_{fis}$, heat deposition per fissioned Am nucleus, $Q/N_{fis}$, the burning rate of Am at the beginning of operation and the amount of Am transmuted in the first year of operation for various target options.

| Material | Initial Am mass (kg) | $N_{fis}$ | $Q/N_{fis}$ (MeV) | dm/dt(t = 0) (g/month) | Burned in first year (kg) |
|----------|---------------------|-----------|-------------------|------------------------|---------------------------|
| $^{241}$Am (a) | 2.58 | 3.00 | 241 | 194 | 1.53 |
| $^{241}$Am + U booster (b) | 2.58 | 3.79 | 229 | 245 | 1.76 |
| $^{241}$Am + U booster + Be reflector (c) | 2.58 | 3.95 | 228 | 256 | 1.79 |

Table 4: Average heat deposition $Q$ and heat deposition per leaked neutron $Q/N$ calculated with MCADS for $^{235}$U, $^{241}$Am and $^{243}$Am targets with the length of 12 cm and radius of 4 cm. All values are given per beam proton.

| Material | $Q$ (MeV) | $Q/N$ (MeV) |
|----------|-----------|-------------|
| $^{235}$U | 766 | 38.1 |
| $^{241}$Am | 1608 | 55.2 |
| $^{243}$Am | 1185 | 48.6 |

Figure 5: Geometry options: bare $^{241}$Am cylinder (option a), $^{241}$Am cylinder covered by a U booster (option b) and $^{241}$Am cylinder covered by a $^{235}$U booster and a $^{9}$Be reflector (option c). The arrows indicate direction of the proton beam.

6. Conclusions

In this paper we have applied the MCADS model based on the Geant4 toolkit for calculating neutron fields and heat deposition in spallation targets containing uranium and americium. We have investigated the criticality of such targets using the Monte Carlo method. We have demonstrated that the critical mass of extended spallation targets containing $^{241}$Am can be evaluated with MCADS by calculating the corresponding neutron multiplication factor as a function of time elapsed since the impact of a beam proton. The MCADS results for the criticality of targets made of $^{241}$Am provide a guideline in the target performance was achieved by the addition of the Be reflector.

The absolute Am burning rates are smaller ($\sim 200$ g/month) than for the simple target ($\sim 500$ g/month). However, the relative burning rates are higher for the advanced options. Indeed, about 42% of the initial Am mass is burned in the first year of ADS operation in the simple pure $^{241}$Am target, as compared with 69% in the advanced target (c).

The distributions of the neutron flux and energy deposition for the options (a)–(c) are shown in Figs. 6 and 7. As seen in these figures, the distributions are more uniform both along the axis of the target and its radius. One can see that adding the booster and the reflector leads to increased average neutron flux by about 50% and 90%, respectively. The highest neutron flux of $4.9 \cdot 10^{16}$ n/s/cm$^2$ is reached locally in the target (c) with its average value of $2.6 \cdot 10^{16}$ n/s/cm$^2$. The highest volumetric energy deposition is below 70 kW/cm$^3$ for the options (b)–(c), which is twice as low as the value of 140 kW/cm$^3$ calculated for the simple $^{241}$Am target.
Figure 6: Distribution of neutron flux inside $^{241}$Am cylindrical target (option a), $^{241}$Am cylinder covered by a $^{nat}$U booster (option b) and $^{241}$Am cylinder covered by the $^{nat}$U booster and a $^{9}$Be reflector (option c). The distributions are calculated for the targets irradiated by 600 MeV proton beam of 10 mA.
Figure 7: Distribution of heat deposition inside $^{241}\text{Am}$ cylindrical target (option a), $^{241}\text{Am}$ cylinder covered by a $^{nat}\text{U}$ booster (option b) and $^{241}\text{Am}$ cylinder covered by the $^{nat}\text{U}$ booster and a $^{9}\text{Be}$ reflector (option c). The distributions are calculated for the targets irradiated by 600 MeV proton beam of 10 mA.
for further studies for their optimization and safe operation in a deep subcritical mode.

Depending on the composition of the target material (pure $^{241}$Am, $^{243}$Am, their mixture or Am$_2$O$_3$), 2.5–4.3 kg of Am can be transmuted into short-lived or stable fission fragments per year of operation in the spallation target of the ADS facility irradiated by 600 MeV proton beam of 10 mA. The results of simulations with targets of different material composition show that the highest rate of americium incineration is achieved in the targets made of pure americium. As demonstrated by simulations, when Am cylinder is covered by the U booster and shielded by the Be reflector, the relative annual incineration rate of Am increases up to 69%. The burning rate may be increased by $\sim 50\%$ if uploading of additional quantities of Am to the spallation target is made on a regular basis. Higher incineration rates can be obtained by increasing the neutron multiplication factor up to $k \sim 0.8$. However, a high energy deposition in these targets creates serious challenges to their cooling systems. Additional studies of technological issues related to the high heat deposition and also to the radiation damage of target materials due to a very high neutron flux predicted in the target are needed to prove the viability of the proposed concept.

In the uranium targets with small admixture of Am long-lived isotope $^{237}$Np is produced following the capture of two neutrons by $^{235}$U and the subsequent beta-decay. However, our estimations show that the amount of $^{237}$Np produced after a year of irradiation does not exceed 100 grams, which is much less than the mass of transmuted MA. The neutron capture on Am nuclei leading to the production of heavier long-lived MA does not change the total amount of MA in the target.

In this paper we did not discuss a well-known solution where a spallation target of a full-scale ADS facility is surrounded by an extended subcritical reactor core [43] [44]. In this case one could not only use neutrons escaped from the spallation target, but also use additional neutrons produced in the reactor core to burn MA placed there. This may significantly, by a factor of 10 or more, increase the amount of burned MA [43] as compared with burning only in the spallation target. Several such ADS facilities can solve the problem of utilization of MA produced in thermal reactors. The thermal energy produced in the spallation target and reactor core can be converted to electricity in order to cover (at least partially) the energy consumed by the accelerator.

Finally, one can note that the highest neutron flux ($4.9 \cdot 10^{16}$ n/s/cm$^2$) is reached in the target with the booster and reflector, and the average neutron flux ($2.6 \cdot 10^{16}$ n/s/cm$^2$) is also high in this target. Therefore, the ADS facilities can be used to study the properties of materials under the impact of intense irradiation by fast neutrons, as well as for basic research.

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