Nuclear Instruments and Methods in Physics Research B 334 (2014) 8–17

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Monte Carlo modeling of spallation targets containing uranium and americium

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ARTICLE INFO

Article history:
Received 22 November 2013
Received in revised form 30 April 2014
Accepted 30 April 2014
Available online 2 June 2014

Keywords:
Spallation reactions
Minor Actinides
Neutron sources
Accelerator Driven Systems
Radioactive waste

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 ^{233,235}U, ^{237}Np, ^{239}Pu and ^{241}Am. Several geometry options and material compositions (U, U + Am, Am, Am_2O_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.

1. Introduction

Many neutrons can be produced in spallation nuclear reactions [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 [3,4] spallation neutron sources and will be used in the facilities to be constructed, e.g., in the ESS project [5]. Such facilities are dedicated to neutron imaging and scattering experiments [6]. Accelerator Driven Systems (ADS) aimed at energy production in subcritical assemblies of fissile materials or burning nuclear waste [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 [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 [10], its role in neutron production is negligible. However, an alternative approach can be also considered to involve fissionable, ^{232}Th, ^{238}U [11], or even fissile, ^{235}U, ^{242m}Am [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 [13], the number of neutrons produced per beam proton is about 3 times higher in a uranium target compared to one made of tungsten, 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 [13] 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,

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http://dx.doi.org/10.1016/j.nimb.2014.04.027
0168-583X/

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 deutrons were used, and the flux of fast neutrons was amplified by a massive uranium sleeve surrounding a non-fissile target [10,17,18].

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 [23–25]. 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 physics 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 abovementioned 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 the 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 NeutronHP 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. The aforementioned lack of accuracy for nucleons with energies below 100 MeV does not affect significantly the results, as such nucleons do not dominate in the considered spallation 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}Am and ^{243}Am, and good agreement with experimental data was obtained. This justifies using MCADS to simulate extended targets containing ^{241}Am and ^{243}Am.

The physics of transmutation of ^{241}Am and ^{243}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}Am and ^{244}Am. After β^--decay with half-life times of 16 h and 10 h these nuclei change finally into long-lived ^{242}Cm and ^{244}Cm, respectively. A sharp rise of the fission cross section at incident neutron energy of $\sim$0.6 MeV leads to the dominance of fission of ^{241}Am and ^{243}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}Am and ^{243}Am very efficiently.

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 [33–36]. 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