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Comparison of different target material options for the European Spallation Source based on certain aspects related to the final disposal

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ABSTRACT

Different target options have been examined for the European Spallation Source, which is under construction in Lund, Sweden. During the design update phase, parameters and characteristics for the target design have been optimized not only for neutronics but also with respect to the waste characteristics related to the final disposal of the target. A rotating, solid tungsten target was eventually selected as baseline concept; the other options considered included mercury and lead-bismuth (LBE) targets suitable for a pulsed source. Since the licensee is obliged to present a decommissioning plan even before the construction phase starts, the radioactive waste category of the target after full operation time is of crucial importance. The results obtained from a small survey among project partners of 7th Framework Program granted by EU 202247 contract have been used. Waste characteristics of different potential spallation target materials were compared. Based on waste index, the tungsten target is the best alternative and the second one is the mercury target. However, all alternatives have HLW category after a 10 year cooling. Based on heat generation alone all of the options would be below the HLW limit after this cooling period. The LBE is the least advantageous alternative based on waste index and heat generation comparison. These results can be useful in compiling the licensing documents of the ESS facility as the target alternatives can be compared from various aspects related to their disposal.

1. Introduction

Various target concepts have been examined for the European Spallation Source (ESS) [1] that is now under construction in Sweden. The decision required consideration of the related environmental issues before the detailed design phase. An operational license can be issued for a facility operating under radiological control only if a feasible strategy for the final decommissioning procedure is presented. Since the highest concentration of radioactivity will be stored in the target during and after the operation of the facility, we focused on this issue. For the comparison of different targets, we consider decommissioning as part of an environmental remediation process that finally ends in an ecological status that is in compliance with any desired later use. In this paper we consider decommissioning only after 40 years of operation as the envisaged lifetime of the ESS facility and 10 years of a cooling-down period. According to the latest design version, the selected target option is a rotating solid tungsten wheel, which can be replaced during the operational phase with a frequency of at least 5 years. The other options considered in the ESS design study included mercury and lead-bismuth (LBE) targets. The eutectic alloy lead-bismuth, tested in the MEGAPIE spallation target [2] at SINQ at the Paul Scherrer Institut (PSI) in Villigen, Switzerland, gives the advantage of lower melting temperatures (melting point 120 °C).

It should be noted that as a part of the ESS Preparatory Phase Project, the possibility of using a Pb-17%Au eutectic (melting point 212 °C) target had also been considered (Pb-Au target LGE), but its applicability was questioned owing to the envisaged corrosion problems in the operational period in addition to its high cost and lack of experience [3].

Further, there is an operating spallation source at the PSI at the MW

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Table 1
Initial elemental composition (weight ppm) of LBE [11].

	B	Mg	Ca	Cr	Fe	Ni	Cu	Ag	Cd	In	Sn	W	Pb	Bi
LBE	1.7	0.2	0.39	0.4	0.2	2.4	2.1	6.6	0.6	28	113	2.7	450000	550000

level with a solid Pb-target (SINQ) in continuous operational mode. Gamma and alpha emitting isotopes after irradiation and cooling were studied for the SINQ target [4–6]. A non-exhaustive list of them is the following: ^{207}Bi, ^{152}Eu, ^{148}Gd, ^{60}Co, ^{154}Eu, ^{90}Sr/^{90}Y, ^{204}Tl, ^{108m}Ag, ^{150}Eu, ^{3}H, ^{158}Tb, ^{101}Rh, ^{146}Pm. However, those results are not applicable to the ESS case due to the difference in power and beam energy. Ghiglino and coworkers [7] investigated a possible Pb target for ESS where the neutronic performances were close to that of the W target. Their water-cooled rotating solid target was considered as an option for the 5 MW ESS project. However, the thermal behaviour of lead for the pulsed source could pose a problem. Since the mechanical model and operational conditions were not available for the authors of this paper the complete isotopic composition of that target for disposal could not be estimated. From the reasons described above, detailed studies were performed only for the three candidate materials for ESS, that is, tungsten, lead-bismuth and mercury target options.

In order to completely meet EU requirements for the decommissioning plans, relevant information concerning primarily the special features of waste generation can be obtained from previous reports, such as the decommissioning plans of the SNS [8] and the MEGAPIE [2] facilities. These plans can be utilized for compiling a preliminary decommissioning plan for ESS. However, the environmental situation (geological, population etc.) as well as the regulations for waste handling and decommissioning of these facilities are different for ESS.

In this paper we consider that the mass and the activation of the structural materials around the target and the accelerator do not differ significantly for the different target options. Therefore our major concern is the final disposal of the target itself.

2. Materials and methods

2.1. Methodology of activity calculation

The radionuclide inventory of the target has a major relevance for reasons of safety, licensing, decommissioning and disposal. While operational key parameters of a neutron spallation target, e.g. the energy deposition and the yield of neutron flux, depend only slightly on the material choice among high-Z elements, the specific composition of the radioisotopes produced depends strongly on it. Even impurities and primary product nuclei might play an important role when the neutron capture cross section is large for the isotope(s) in question. From the nuclide inventory the waste index *WI* (often referred to as clearance index *CI*, see Eq. (1) below) is derived. This value is related to the similar, but less practical term of “radiotoxicity” which is the fictitious collective dose consequence of a waste inventory. Another quantity, which can be derived from the nuclide inventory, discussed further in this paper, is the decay heat. It is an important factor for the cooling requirements during transient phases of the operation, which might be short interruptions or a few weeks for changing the target.

Due to the complexity of the geometry and material compositions involved, the calculation of the nuclide inventory requires particle and radiation transport codes. For the calculations reported here, the Monte Carlo program MCNPX [9] was used covering the transport of primary protons and generated particles in a wide range of energies. Since the aim of the neutron spallation source is the production of low-energy neutrons starting from the high-energy spallation reactions, MCNPX has been chosen as the best validated code. In most of the calculations, the initial proton beam energy EP is 1.334 GeV. This energy has been proposed for ESS in an early design study [10]. However, the final energy of 2 GeV is foreseen in the future for the ESS project in Lund. This is the new aim presented at Technical Design Report [1].

Since the nuclear production reaction cross sections are almost linear with energies above 300 MeV, the expected changes in the nuclide inventories are comparable. Due to the higher energy transferred to the nucleus even more neutrons will evaporate. This will cause a broadening of the distribution of isotopes produced. The dip clearly seen for lower energies at around Z = 50 and A = 120 in the charge and mass distribution of the residual nuclei from heavy mass targets will disappear.

2.1.1. LBE

For LBE target the geometry has been taken from a FZJ report [11] and also used for the studies of Clausen et al. [10] The target vessel was 1 m long, but the proton beam of E_P = 1.334 GeV would be stopped already after 89 cm in LBE (density ρ = 10.3 g/cm^3 at 200 °C operation temperature). The beam profile of the proton beam is assumed to be of a Gaussian shape with standard deviations σ_x = 3.35 cm and σ_y = 1.00 cm. The distribution was cut at 3σ.

The initial material composition for LBE is shown in Table 1. The major components (lead and bismuth) are represented by their nominal concentrations. The contributions of impurities were taken from a material analysis of LBE used for the MEGAPIE project. Implications on the nuclide inventory will be discussed below. For the calculation of the nuclide inventory, a modified version of MCNPX2.5.0 [12] was applied. This modified code allows that all elements of the material composition, including impurities, undergo nuclear reactions at each collision (subsequently the appropriate weight is applied). This is of advantage if reasonably small statistical uncertainties also for isotopes produced from impurities are required. To calculate the nuclide inventory generated by low-energy neutrons, fluxes for neutrons less than 20 MeV are first obtained. For all other reactions, production rates of the residual nuclides are collected. The neutron fluxes are folded by external programs like SP-FISPACT07 [13] and CINDER90 [14], version 7.4.2, with neutron production cross sections taken from EAF2007 [15] and the CINDER library, respectively. Both programs also account for the build-up and decay during irradiation; thus, the change of the initial material composition is considered as well. The coupling between MCNPX and the build-up and decay programs is done by a PERL script [16]. In the present calculation the evaluated cross section tables ENDF/B-VI are used for neutron energies lower than 20 MeV and physical models otherwise. To probe the sensitivity of the chosen model, the calculation was done with two different physical models. To describe the first step in the reaction mechanism, the spallation process, intranuclear cascade (INC) models, the Bertini code [17,18] and INCL4.2 [19] were applied. For the de-excitation of the nucleus the Bertini model is coupled to the code of Dresner [20] and Atchison [21] (RAL) and INCL4.2 – to the abrasion-ablation model ABLA [22]. These options allow for fission of the nucleus and evaporation of neutrons, protons, and light isotopes up to ^{4}He. Since the ABLA version implemented in MCNPX2.5.0 cannot describe the emission of ^{3}H, the predicted amount of tritium is much too low.

2.1.2. Mercury target

Activity assumptions for mercury are based on respective work for SNS [23], Eurisol [24] and former ESS studies [25,26]. However, because of discrepancies with some relevant radiotoxic nuclides, some calculations were performed using MCNPX/CINDER90 in combination with Isabel-ABLA, except for tritium, where the Bertini-Dresner