Radiation protection calculations related to the decommissioning of the European Spallation Source

Booklet of thesis

by

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Introduction

For the sake of the future generations, decommissioning planning and providing the necessary resources to be at hand in time are required for medical, industrial and research facilities producing radioactive waste [1]. Planning is important to ensure that decommissioning will be carried out in a safe and effective way. Several types of information are required to make a precise decommissioning plan. For proper planning, a good estimation of the radioactive inventory of the facility is needed[2]. The compilation of an initial decommissioning plan is obligatory before obtaining the construction license in order to optimize the design in respect of the decommissioning. The decommissioning plan needs to be reassessed every few years.

The European Spallation Source (ESS)[3] is under construction in Lund, Sweden. It will be the world's most intense neutron source suitable for research related to materials sciences, nanotechnology, energy, health, and environment. The ESS includes a linear proton accelerator, a target monolith building with a rotating tungsten target, neutron instruments and experimental laboratories. The ESS will be the world's first spallation source using long pulses in the ms range, implementing the ideas of Ferenc Mezei[4]. The 2 GeV protons impinge on the solid, rotating tungsten target wheel and initiate spallation reactions, releasing neutrons from the tungsten nuclei. After moderation, the neutrons are guided to the experimental rooms. The tungsten target is planned to be replaced in every five years. The envisaged lifetime of the ESS is 40 years.

During the operational time the structural materials will be activated resulting in radioactive waste at the time of their replacement and/or decommissioning. In order to decrease the hazard represented by the radioactive waste produced during ESS operation, preliminary analysis of the activation is required. The material selection and the design can be optimized based on these calculations. In the initial decommissioning plan for the ESS[5] we identified the most crucial parts of the facility regarding to the radioactive waste.

Objective

The aim of this PhD thesis is to present calculation methods developed for predicting the quality and the quantity of the produced radioactive waste at ESS thus in turn supporting the material selection and design. The results show that in addition to the bulk material composition impurities also have a significant effect on the activation. The developed calculation methods are able to identify the source elements of the most important radionuclides in terms of decay gamma dose rate (for maintenance and refurbishment) and waste index (for processing requirements and for temporary and final disposal). Based on these calculations, the material selection can be optimized for decommissioning. The object of this thesis focused on the most crucial parts of ESS specified in the following; however, these methods can be extended for the whole facility.

The rotating solid tungsten target will apparently be the most active part. The activation of the neutron guides and inserts is an important issue, because the replacement of the guides is envisaged after a few years use due to radiation damage, mechanical failure, and progress in guide technology. The beryllium reflector is also envisaged to be replaced and the activated beryllium requires specific treatment. At the ESS, metal-based shielding plays an important role in the attenuation of high-energy neutrons. The activation of the materials in experimental caves needs particular consideration in terms of radiation protection of the workers. In this thesis the NMX macromolecular diffractometer experimental cave was studied as an example of instrumentation. The activation properties in these selected parts depend strongly on the material choice.

In order to validate the model calculations, aluminium and glass neutron guide substrate samples were irradiated at the Budapest Neutron Centre and the measured activity concentrations were compared with the model predictions.

Applied methods

1. Methodology of the activity calculation

Due to the complexity of the geometry and material compositions involved, the calculation of the nuclide inventory is performed with particle and radiation transport codes. For the calculations reported here an appropriate version of the Monte Carlo program MCNPX was used covering the transport of primary protons and generated particles in a wide range of energies. The radionuclide inventory was calculated with MCNPX2.7 [6] using ENDF/B-VII cross-section libraries coupled with CINDER'90 [7] nuclear inventory code. In case of the target, neutron guide, inserts, and beryllium reflector, the geometry used in calculations was defined as it was given in ESS TDR[3]. The target wheel diameter is 2.5 m and it consists of 33 sectors. The proton beam energy is 2 GeV and the time-average current is 2.5 mA which means 1.56×10^{16} proton/s time-average operational intensity. For the activation calculations of NMX materials and metal-based shielding MCNPX models were compiled using the neutron beam as the input. In order to prove the sensitivity of the chosen model, the MCNPX calculations were performed with different spallation models.

2. Methodology of waste categorization

For waste classification we used the method that is based on the definition of negligible dose which is associated with a risk that is too low to justify any countermeasures[8]. This method defines a waste index *WI* for the whole waste stream which directly relates the waste classes to the respective values of clearance level (*CL*) associated with the negligible dose. Clearance levels are published in IAEA publications and EURATOM directives, but the most precise compilation of limiting scenarios and clearance levels is in RP122[9]. Several radioisotopes originating from proton activation and spallation do not appear in former compilations[9,10,11] as these publications were elaborated primarily for neutron-rich radioisotopes generated in nuclear fission reactors. Two methods for the estimation of the missing clearance levels were applied for the generation of overall waste indices. In both cases the components of the effective dose or dose rate were

compared with those of a well-known standard radionuclide. External exposure scenarios in case of gamma-emitting radionuclides, various internal exposure pathways in case of alpha- and beta-emitting radionuclides, were used to define the effective dose and thus the estimate of the most restrictive *CL*.

3. Irradiation experiments

Aluminium and glass neutron guide substrate samples were irradiated at research reactor of the Budapest Neutron Centre. The nominal material composition was verified by X-ray fluorescence spectroscopy (XRF). The activity concentrations of the irradiated samples were measured by gamma spectroscopy. The irradiation experiments were simulated using the MCNPX model of the reactor core and CINDER'90 activation code. The results of the experiments and simulations agreed within the margin of estimated errors.

Thesis statements

1. As the waste index is obliged to cover all artificial radionuclides present in the material in question I defined the clearance level (*CL*) for those components of the ESS tungsten target inventory which were missing from previous *CL* compilations[9,10,11] by setting up scenarios for estimating *CL*s similarly to the procedures described in the mentioned compilations [T1].

2. I assessed the radioactive inventory and waste index of the ESS tungsten target in comparison with other previously studied target options.

a. I compiled different compound high energy models combining MCNPX2.7[6] and CINDER'90[7] calculation tools in order to assess the inventory and consequently the waste index of the ESS tungsten target. Spallation events were modelled in two ways: in two separate steps or in a single procedure. The first step of the reaction mechanism of the proton-induced spallation process was described with intranuclear cascade (INC) models: Bertini[12], INCL4.2[13], and Isabel[14] codes were applied. For the adequate description of the de-excitation of the nuclei, that constitutes the second step of the process, these models were coupled to Dresner[15] and ABLA[16] codes, respectively. The CEM02[17] model that was also applied in the calculations included these two consecutive steps of spallation process modelling in one package. The runs with different combinations of these models gave approximately the same total activity concentration for the target without considering the tritium content. The resulting value was 1.7×10^{9} Bq/cm^3(±10%) using the different models; this confirmed their adequacy for our purpose. However, in case of tritium production the differences of the applied models were of 3 orders of magnitude; the CEM02 combined spallation model gave the most conservative estimation of tritium production: 1.4×10^{10} Bq/cm^3. Except for tritium production, the combination of INCL4.2 and ABLA codes resulted in the most conservative estimation of the relevant isotopes contributing significantly to the waste index [T1, T8, T11, T14].

b. Based on the calculation of radioactive inventory I showed that the tungsten target would be classified as high level waste (HLW) after 5-year irradiation and 10-year

cooling time. The inventory was estimated using 2 GeV incident proton energy and activity concentrations were defined after 5 years of irradiation and 10 years of cooling time; this resulted in a waste index of 3.2×10^{7}. I showed that the tungsten target has a more favourable waste index than previously considered lead-bismuth eutectic (LBE) and mercury targets assuming similar operational conditions. The nuclides having the greatest share in the compound waste index are ^{148}Gd, ^{133}Ba, ^{152}Eu, ^{179}Ta, ^{60}Co, and ^{154}Eu. I determined that 99% of ^{60}Co production is due to cobalt impurity. Therefore, I advised to use structural materials with low cobalt content [T1, T8, T12].

3. a. The possible neutron guide substrate materials located the closest to the target were investigated for radiation safety during operation and decommissioning. Coupling MCNPX2.7[6] to CINDER'90[7] code I estimated the radioactive inventory of three different candidate materials as potential ESS neutron guides. In order to assess the radiation hazard of disassembling the device I showed that after 5 years of operation the aluminium guides (1.9×10^{7} Bq/cm^3) would be less active than the float (1.1×10^{8} Bq/cm^3) and zerodur (1.2×10^{8} Bq/cm^3) guides. However, in case of zerodur, the tritium gives the main contribution. The main gamma-emitting isotopes that define the external dose exposure of the personnel performing the replacement of the device are ^{65}Zn in case of zerodur and aluminium, and short-lived ^{24}Na in case of float. Therefore, I advised to decrease the amount of zinc in aluminium and zerodur substrate materials [T6, T7, T10, T12].

b. I studied the effect of material selection on the radionuclide inventory of metal-based shielding blocks by assessing radioactivity generation applying the method of coupling MCNPX2.7[6] to CINDER'90[7] code thus providing neutron activation conditions in terms of energy flux and energy distribution. Three materials were examined: iron, copper, and aluminium. Immediately after shutdown the steel shielding block will be the least active, the dominant radioisotopes being ^{55}Fe, ^{59}Fe, ^{54}Mn, ^{56}Mn. Therefore, it is advised to decrease the manganese content of the steel without changing its advantageous thermal and physical behavior. After 5 days of cooling time, the copper shielding block becomes the least radioactive, because in the activated copper shielding short-lived copper isotopes ^{64}Cu, ^{66}Cu dominate. In activated aluminium shielding short-lived ^{28}Al, ^{56}Mn, ^{64}Cu, and long-lived ^{65}Zn will dominate during the maintenance and replacement operations; this

requires to decrease the amount of zinc in the material composition in facility parts where aluminium is applied [T4, T13].

4. I performed activation and dose calculations to estimate the dose consequences of the handling of the activated beryllium reflector. In order to assess the radionuclide inventory simulations were performed with MCNPX2.7[6] using ENDF/B-VII cross section libraries coupled with CINDER'90[7] similarly to the method of statement 3. I showed that at shutdown after 1-year operation the most active gamma-emitting isotopes are ^{56}Mn (1.8×10^{9} Bq/cm^3) and ^{7}Be (9.7×10^{8} Bq/cm^3). The dose rate on the external surface of the beryllium reflector after 1-year irradiation time is 720 Sv/h at the end of operation, decreasing to 60 mSv/h after 2 weeks of cooling time. Owing to this dangerous level I recommended to store the beryllium in hot cell before final disposal during the decommissioning process. It is recommended to wait at least two weeks of cooling time before the replacement of the beryllium. After two weeks of cooling time the most active gamma emitters in the activated beryllium are ^{7}Be, ^{51}Cr, ^{46}Sc, ^{95}Nb, ^{95}Zr, ^{60}Co, and ^{88}Y. It means that the quantity of cobalt, chromium, scandium, niobium, zirconium, and yttrium impurities should be decreased as much as possible [T5, T9].

5. Irradiation experiments were designed and carried out for the purpose of validating the simulation procedure performed with the combination of software tools introduced above. Aluminium neutron guide substrate samples were irradiated for 6 hours at one of the fast neutron channels at the research reactor of Budapest Neutron Centre. Using the nominal composition of the samples the simulations showed a rather big difference from experimental results. Based on these results, I recommended that the nominal composition of the sample (with special respect to trace elements) should be verified by chemical analysis. In turn, using the material composition obtained with XRF analysis the simulations resulted in the same order of magnitude as experimental data. After 1 week of cooling time the most active isotopes in two different aluminium samples were ^{65}Zn (4.8×10^{4} and 7.1×10^{7} Bq/cm^3), ^{64}Cu (2.4×10^{4} and 1.8×10^{6}Bq/cm^3) and ^{51}Cr (3×10^{6} and 2.39×10^{6}). These results show that the aluminium substrate should contain the lowest possible concentrations of Zn, Cu, and Cr. Considering the discrepancies between the nominal and re-determined elemental composition of the samples, the agreement between

the measured and calculated results was sufficient to confirm the suitability of the combined simulation procedure presented in the thesis. This means that the calculations performed with the same software tools for predicting the activation of the examined substrates under the assumed ESS irradiation conditions can also be considered accurate enough. In case of ^{60}Co the measured data were one order of magnitude larger than the simulated one. It is probably due to the fact that cobalt content was not detected by XRF and not available in the nominal composition. Therefore, I advised to validate the material compositions using different experimental techniques beside XRF, e.g. activation analysis [T3].

6. I performed activation calculations for materials of ESS NMX experimental cave considering the walls assumedly made from ordinary concrete and the floor that was scheduled to be manufactured from aluminium. An adjusted version of the previously described combination of MCNPX2.7[6] and CINDER'90[7] was applied, the former code used the ENDF/B-VII cross-section libraries as before. I showed that after 40 years of operation the major contributor for the total activity defining the dose field for the operators that would enter the area was the aluminium false floor (2.46 kBq/g) due to the production of ^{28}Al and ^{56}Mn. Therefore, I recommended to ignore aluminium as a structural material and to use other, preferably nuclear grade materials or steel covered boron containing floor. The main component of the concrete wall structure responsible for its contribution to the dose field will be ^{24}Na during operations and interim maintenance and refurbishment procedures [T2].

Related publications

[T1] Zs. Kókai, Sz. Török, P. Zagyvai, D. Kiselev, R. Moormann, E. Börcsök, L. Zanini, A. Takibayev, G. Muhrer, R. Bevilacqua, J. Janik: Comparison of different target material options for the European Spallation Source based on certain aspects related to the final disposal, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, Volume 416, 1 February 2018, Pages 1-8 https://www.sciencedirect.com/science/article/pii/S0168583X17309977

[T2] G. Náfrádi, Zs. Kókai, P. Zagyvai: Az NMX mérőhelyének sugárárnyékolása (Shielding of ESS NMX cave), Nukleáris Technikai Szimpózium, 2017

[T3] Zs. Kókai, E. Dian, L. Zanini, F. Mezei, A. Takibayev, E. Klinkby, Sz. Török, P. Zagyvai, G. Patriskov, F. Gergely: Neutron guide activation calculations for the European Spallation Source and validation measurements at Budapest Neutron Centre, International Collaboration on Advanced Neutron Sources (ICANS XXII), 27-31 March 2017, Oxford, UK, http://icansxxii.iopconfs.org

[T4] D. Dijulio, C. P Cooper-Jensen, H. Björgvinsdóttir, Zs. Kókai, P. M. Bentley: High-energy in-beam neutron measurements of metal-based shielding for accelerator-driven spallation neutron sources, Physical Review Accelerators and Beams, 19, 053501 (2016) https://journals.aps.org/prab/pdf/10.1103/PhysRevAccelBeams.19.053501

[T5] Yongjoong Lee (Ed.): ESS Materials Handbook, Beryllium dose maps in the target area, ESS-0028465, 2015

[T6] Zs. Kókai, L. Zanini, F. Mezei, A. Takibayev, E. Klinkby, P. Zagyvai, Sz. Török: Activation of the neutron guides designed for the European Spallation Source, 6th International Conference "Charged & Neutral Particles Channeling Phenomena - Channeling 2014", Capri, October 5-10, 2014

[T7] Zs. Kókai, L. Zanini, F. Mezei, A. Takibayev, E. Klinkby, P. Zagyvai, Sz. Török: Neutron guide activation and handling for the European Spallation Source, ICANS XXI: International Collaboration on Advanced Neutron Sources, Mito, 29 Sep - 3 Oct 2014

[T8] Kókai Zs., Zagyvai P., Török Sz.: Az Európai Neutronkutató Központ leszerelési terve (Decommissioning plan of ESS), Nukleon VII.évf. 4.szám, 2014, http://nuklearis.hu/nukleon/az-europai-neutronkutato-kozpont-leszerelesi-terve

[T9] P. Zagyvai, Zs. Kókai, D. Ene: Initial decommissioning plan for ESS, SSM2012-131, 2013

[T10] Zs. Kókai, P. Zagyvai. Sz. Török, L. Zanini: Health physics calculations related to the target station radiation protect, Accelerator and Integrated Control System Retreat, 2013

[T11] Zagyvai P., Kókai Zs.: A nukleáris energiatermelés radioaktív hulladékai - lektorált egyetemi jegyzet (Radioactive waste of nuclear fuel cycle), 2013 https://www.scribd.com/doc/316374945/Nuklearis-Uzemanyagciklus-Radioaktiv-Hulladekai-MTA-EK

[T12] Zagyvai P., Kókai Zs., Török Sz.: Leszerelési tervek európai nagyberendezésekhez (Decommissioning plan of European research facilities), Nukleáris Technikai Szimpózium, Budapest, 2013

[T13] Kókai Zs., Zagyvai P., Bodor K.: Az Európai neutronkutató központ (ESS) előzetes leszerelési tervével kapcsolatos kutatások (Research related to initial decommissioning plan of ESS), XXXVIII. Sugárvédelmi Továbbképző Tanfolyam, Hajdúszoboszló, 2013

[T14] Kókai Zs., Breitner D., Török Sz.: Nagyaktivitású hulladék végleges elhelyezésére vonatkozó nemzetközi elképzelések (Final disposal of high level radioactive waste), Nukleáris Technikai Szimpózium, Paks, 2012

References

[1] Decommissioning of medical, industrial and research facilities, IAEA Safety Standards Series no. Ssg-49, Vienna, 2019

[2] Methodologies for assessing the induced activation source term for use in decommissioning applications, Safety Reports Series No. 95, IAEA, Vienna, 2019

[3] S. Peggs (Ed.), ESS Technical Design Report, 2013.

[4] F. Mezei, Long pulse spallation sources, In Physica B: Condensed Matter, Volumes 234–236, 1997, Pages 1227-1232

[5] P. Zagyvai, Zs. Kókai, D. Ene: Initial decommissioning plan for ESS, SSM2012-131, 2013

[6] D. Pelowitz. (ed.), 2011. MCNPX User's Manual, Version 2.7.0, LA-CP-11-00438

[7] W. B. Wilson, S. T. Cowell, T. R. England, A. C. Hayes & P. Moller, A Manual for CINDER'90 Version 07.4 Codes and Data, (2007)

[8] Radiobiology for the Radiologist" E. J. Hall & A. J. Giaccia. 6th ed. Lippincott. Williams & Wilkins, Philadelphia 2006

[9] Radiation Protection 122/I *Guidance on General Clearance Levels for Practices* Recommendations of the Group of Experts (2000)

[10] Radiation Protection 157 *Comparative Study of EC and IAEA Guidance on Exemption and Clearance Levels* Directorate-General for Energy Directorate D — Nuclear Energy Unit D4 — Radiation Protection (2010)

[11] IAEA Safety Reports Series 44 Derivation of Activity Concentration Values for Exclusion, Exemption and Clearance Vienna, 2005

[12] H.W. Bertini, Phys. Rev. 188 (1969) 1711–1730

[13] A. Boudard, J. Cugnon, S. Leray, and C. Volant., Phys. Rev. C 66, 044615 (2002)

[14] Y. Yariv, Z. Fraenkel, 1981. Phys. Rev. C. **24**. 488-494

[15] L. Dresner, 1981. EVAP-A Fortran Program for Calculating the Evaporation of Various Particles from Excited Compound Nuclei, Oak Ridge National Laboratory report ORNL-TM-7882

[16] A. R. Junghans, M. de Jong, H.-G. Clerc, A. V. Ignatyuk, G. A. Kudyaev, K.-H. Schmidt, Projectile-fragment yields as a probe for the collective enhancement in the nuclear level density, In Nuclear Physics A, Volume 629, Issues 3–4, 1998, Pages 635-655

[17] K. K. Gudima, S. G. Mashnik, V. D. Toneev, Cascade-exciton model of nuclear reactions, Nuclear Physics A, Volume 401, Issue 2, 1983, Pages 329-361