HORIZON 2020

HORIZON 2020 RESEARCH AND INNOVATION FRAMEWORK PROGRAMME OF THE EUROPEAN ATOMIC ENERGY COMMUNITY

Nuclear Fission and Radiation Protection 2018 (NFRP-2018-4)

Project acronym:	**SANDA**
Project full title:	**Solving Challenges in Nuclear Data for the Safety of European Nuclear facilities**
Grant Agreement no.:	**H2020 Grant Agreement number: 847552**

Workpackage N°:	***WP5***
Identification N°:	***D5.4***
Type of document:	***Deliverable***
Title:	***Contribution to Nuclear data needs for HLW disposal from KIT***
Dissemination Level:	***PU***
Reference:	
Status:	***VERSION 1***
Comments:	….

	Name	Partner	Date	Signature
Prepared by:	R. Dagan	16	3.12.2021	Ron Dagan
WP leader:	R. Jacqmin	3	30.09.2022	JACQMIN Robert 116318 Signature numérique de JACQMIN Robert 116318 Date : 2022.09.30 14:51:54 +02'00'
IP Co-ordinator:	E. González	1		Signed by E. Gonzalez el 17/10/2022 with a certificate emitted by AC Sector Público

Abstract

KIT contribution to task 5.1 aims on evaluating the nuclear data needs for HLW (High Level Waste) disposal. The main activity focuses on studying the impact of nuclear data on decay heat and biological hazards via nuclei, which have considerable high geochemical mobility. Further topics concern models on neutron backscattering from the walls surrounding the HLW casks within a repository, which in return affects strongly the ambient dose rate.

For all those topics, the most relevant isotopes were identified. For simulation tools and in particular for the secondary scattering, the importance of the angular distribution is pointed out. Some drawbacks of the common methods, mainly in the higher keV range, are discussed and further investigation and improvement suggestions, among others in view of foreseen dedicated measurements, are introduced.

The issue of the uncertainties of the nuclide inventory due to existing nuclear data is beyond the scope of this work within Task5.1 and it introduced in a general form within Task 5.2.

Content

1. Introduction

Safe procedures for the disposal of HLW (High Level Waste), either in the form of spent nuclear fuel (SNF) elements or of reprocessed vitrified waste, are of crucial importance for the acceptance of the entire nuclear energy program worldwide. The fact that, only in January 2022 the first final repository in Sweden has been officially approved shows the complexity of the issue. Moreover, the diversity of solutions proposed and at different countries call for a more deeper understanding of the nuclide inventory together with the type of galleries in which they should be disposed. For example, in Finland and in Sweden, the underground disposal facilities are at a final stage. The host rock is granite, which raises waste container corrosion issues due to the possible diffusion of water, which is a potential carrier of radioactive, mobile fission product nuclides. On the other hand, in Germany, there is the availability of salt and clay rock, which has advantages as far as the mobility of the disposed radioactive nuclides is concerned. However, the necessary research work on the integrity of the spent fuel assemblies or waste containers in view of their radioactivity, chemical and thermal properties is still ongoing. In this sense, the nuclear data information needed to characterize the type of waste, its quantity, radiotoxicity type, decay time and chemical behavior and mobility in soil air or water is of a crucial importance for designing a safe and acceptable final repository. In addition, the fact that in several European countries the final repositories approval might not be completed in the next decades means that the existing temporary storage of the fuel in casks might exceed 50 to 100 years. Consequently, the decay heat, radiation damage, chemical phase of the fission- and activation products and their mobility, all call for an accurate nuclear data knowledge and property handling to ensure a safe dry storage over long periods.

This study is a contribution to the above research on three nuclear data related topics. The first one concerns mainly the decay heat of fuel assemblies in the first stages after withdrawal from the reactor core, cooling and transfer to dry storage. The second topic concerns the need for an accurate definition of important nuclides, emphasizing the highly mobile ones in view of their half-life time and consequently their radioactive biological hazard. The third topic concerns the needs for nuclear data for the final disposal site. In the latter case, the importance of neutron scattering processes from the standpoint of radiation protection within the site galleries is shown.

2. Decay Heat

The decay heat importance got its full "recognition" with the three main accidents in nuclear facilities:

- Three-mile Island: March 1979,
- Chernobyl: April 1986,
- Fukushima: March 2011.

All three events are connected to the decay heat issue. In the first and third cases, decay heat was directly responsible for the heat which caused the damage. In Chernobyl, an experiment dedicated to investigate the feasibility of mitigating the decay heat vulnerability of nuclear reactors ended up in the worst nuclear scenario.

In all three cases, the decay heat is sufficiently well approximated by the known Borst Wheeler formalism [1]:

Equation (1)

$$P(t,T) = \frac{2.7P_0}{200\times 1.6\times 10^{-13}}\int_t^{t+T} dt'\, t'^{-1.2} = \frac{13.5P_0}{3.2\times 10^{-11}}\left[t^{-0.2} - (t+T)^{-0.2}\right] MeV/\sec$$
$$= 4.1\times 10^{11} P_0\left[t^{-0.2} - (t+T)^{-0.2}\right] MeV/\sec$$
$$= 0.065 P_0\left[t^{-0.2} - (t+T)^{-0.2}\right] Watt$$

Where the Borst Wheeler function definition contains:

P (*t,T*) - Power emitted at time *t* after shutdown of a reactor which operated for time *T* before shut down.

In spite of the simplicity of this expression, this approximation is very good in the range $1< t<10^6$ sec and, in several cases, good enough up to few years. Clearly, it is sufficient to demonstrate the evident risk immediately after shut down and the mandatory redundant emergency systems to avoid scenarios like the one in Fukushima.

The worst Wheeler functions cannot be applied for longer periods than 1-2 years, in which, the decay heat is an important factor for the determination of the storage conditions of burned fuel. Here for, one distinguishes 3 main situations:

- Pool storage for short time cooling
- Temporary Dry storage
- Final disposal in dedicated repositories.

2.a Pool storage for short time cooling

The incident in the wet storage in Fukushima in which the fuel assemblies were melted and damaged emphasized the need of an accurate evaluation of short-lived isotopes and their half-life times.

At first observation, the generated decay heat could have been approximated by the above shown equation (1) and thus the necessary protection to the pool could have been taken and the unfortunate spent fuel melting scenario might have been avoided. The fact that it occurred evidently demonstrates the need for a more extensive analysis of the exact nuclide vector of the spent fuel, in particular the short and mobile radioactive materials, which can obviously cause a meltdown of the fuel, even after one year of cooling, as happened in Fukushima. The above Borst Wheeler approach is obviously not sufficient.

An accurate estimation of decay heat as a function of time after irradiation is obtained mainly by Monte Carlo codes which are connected to burn up codes from which the nuclide dependent decay heat can be determined. As far as the decay heat is concerned, one has to assure that the important isomers are accounted for. In view of the fact that some codes do not handle isomers one should, as appropriate, separate the flux solver from the depletion code and assure that the isomers are included. An important example is the Ba137m, which is a decay product of Cs137 and contributes considerably to the decay heat.

The nuclides, which have a particular large contribution to the decay heat, and thereafter their nuclear data, should be examined carefully. They are in general divided into time groups as a function of the time elapsed after shut down. It is clear from equation (1) that for the Fukushima accident for the first hours, the emergency cooling system within the reactor should had been able to remove at least 6.5% of the initial fission power in the first hours, and down to 2% of the initial heat after 2-3 days, making a deeper analysis on particular isotope decays useless. The impact of specific decaying isotopes rises up only after the short–lived isotopes have died away.

In this section the residual relevant isotopes are shown from 6 months to several tens of years where an improved analysis of some isotopes could motivate changing the handling procedure of spent fuel at temporary storage and at the permanent disposal facilities.

In this work, relevant characteristic calculations were performed for a typical theoretical PWR operating up to 55MWd/kg heavy metal. The calculations employed the standard option of the NUCLEONICA Program [2] for the spent fuel inventory. The list of isotopes in Table 1 presents more of 98% of the nuclides contributing to the remaining heat and therefore are the main elements to be looked at.

Four isotopes have a very short half-life and it is clear that their precursors should be examined too. Pr144 with a half-life time of 17.28 min is a decay product of Ce144, which appears also on the list and has thereafter a particular importance, contributing about 25% to the total generated heat.

Another important nuclide is Rhodium Rh106 with a half-life time of 30 s. Its precursor is Ruthenium Ru106 with a half-life time of 1.02015 y. At such time after shut down, the decay

of Ru106 is of crucial importance and, through the daughter decay of Rh106, accounts for about 22 % of the whole generated heat.

The third isotope is the isomer Ba137m. This is a daughter product of Cs137. Its importance is not that big as the contributions of the above mentioned two isotopes, as it represents only about 4% of the heat at this period of time. Nevertheless, the Cs137 has a half life time of about 30 y and is, through Ba137m, the main contributor to the heat production at later time periods as will be discussed below.

The same arguments mentioned above for Cs137 are also valid for Y90 and its precursor Sr90. Again, as for Cs137, the Sr90 has a half-life time of about 28 years, which increases its importance at longer decay times.

Table 1: Heat generated by the most dominant nuclides (98.2% of the generated heat) 6 months after reactor shut down for a representative 55 MWd/kg_{HM} burnup.

Z	nuclide	half time	decay heat (W)
59	Pr144	17.28 m	1.074e+5
45	Rh106	30. s	1.016e+5
55	Cs134	2.06509 y	5.346e+4
41	Nb95	34.9907 d	4.007e+4
96	Cm242	162.928 d	3.597e+4
40	Zr95	64.0324 d	2.148e+4
56	Ba137m	2.552 m	1.243e+4
39	Y90	2.67083 d	1.220e+4
58	Ce144	285. d	9.607e+3
39	Y91	58.5104 d	8.083e+3
96	Cm244	18.0004 y	7.068e+3
94	Pu238	87.698 y	4.770E+3
44	Ru103	39.2604 d	4.699e+3
38	Sr89	50.5706 d	4.078e+3
55	Cs137	30.0406 y	3.754e+3
63	Eu154	8.59302 y	3.519e+3
38	Sr90	28.7898 y	2.273e+3

The calculations for the same operational conditions as above were repeated for a cooling time of 2 years. In this case Table 2 shows the growing importance of Cs137 and Sr90 whilst the isotopes with half-lives of up to a few months like Sr89, Ru103 etc. die away. Table 2 presents the spent fuel vector after 2 years for elements contributing about 98.2% of the heat

produced, where indeed the contributions of Sr89, Ru103 are marginally small and the nuclides are neglected.

Table 2: Heat generated by the most dominant nuclides (98.2% of the generated heat) 2 years after reactor shut down for a representative 55 MWd/kg_{HM} burnup.

Z	nuclide	half time	decay heat (W)
45	Rh106	30 s	3.668e+4
55	Cs134	2.06509 y	3.231e+4
59	Pr144	17.28 m	2.835e+4
56	Ba137m	2.552 m	1.201e+4
39	Y90	2.67083 d	1.177e+4
96	Cm244	18.0004 y	6.671e+3
94	Pu238	87.6984 y	4.865e+3
55	Cs137	30.0406 y	3.627e+3
96	Cm242	162.928 d	3.502e+3
63	Eu154	8.59302 y	3.117e+3
58	Ce144	285. d	2.534e+3
38	Sr90	28.7898 y	2.192e+3
51	Sb125	2.758 y	8.626e+2

2.b Towards temporary Dry storage

Past 5 years of decay time, Cs137 and Sr90 become the most dominating elements as far as the generated heat is concerned. One should point out that the amount of heat produced in this time domain is essential for taking decisions about the type of cooling needed for pools with forced flow and/or pools with natural flow. The better the decay heat can be estimated, the sooner the elements can be removed to dry storage.

In a Benchmark calculation lead by SKB Sweden [3] the nuclides which contribute to the decay heat were isolated and analysed by several codes. The importance the main isotopes can be seen for five cases, from 4 years, up to 22 years of cooling time, in Table 3.

The pairs Cs137/Ba137m and Sr90/Y90 generate about 40% up to 61% of the decay heat at the time range 4.5 to 21.5 y respectively. Again, as shown by the former calculation, the contribution of Cs134 (half-life time 2y) vanishes in this period as well as the contribution of Ru106.

Table 3: The main heat decay generators between 4.5 and 20 years cooling time based on the SKB Benchmark [3]. The percent number near each isotope emphasizes the partial contribution of heat of only one specific isotope to the total heat produced at a certain time after shut down.

Case1: ~4.5 y decay time	Case2: ~8.5 y decay time	Case3: ~10 y decay time	Case4: ~13.5 y decay time	Case5: ~21.5 y decay time
134Cs 21%	137Cs/ 137mBa 29%	137Cs/ 137mBa 31%	137Cs/ 137mBa 29%	137Cs/ 137mBa 32%
137Cs/ 137mBa 20%	90Sr/90Y 26%	90Sr/90Y 29%	90Sr/90Y 26%	90Sr/90Y 29%
90Sr/90Y 19%	244Cm 15%	244Cm 13%	238Pu 18%	238Pu 14%
106Ru/ 106Rh 11%	238Pu 12%	238Pu 11%	244Cm 13%	244Cm 12%
244Cm 10%	134Cs 8%	134Cs 5%	241Am 7%	241Am 9%
238Pu 7%	241Am 4%	241Am 5%	154Eu 2%	240Pu 2%
144Ce/ 144Pr 6%	154Eu 3%	154Eu 2%	134Cs 1%	154Eu 1%
154Eu 2%		240Pu 1%	240Pu 1%	
141Am 1%				

After 20 years, the importance of the decay heat has direct impact on the geological disposal beyond the interim storage. As far as the isotopes is concerned, the minor actinides become dominant. Thereafter, in Table 4, heat data obtained with NUCLEONICA for Tables 1 and 2, are given at 40 years cooling time.

The pairs Cs137/Ba137m and Sr90/Y90 generate about 50% of the heat. However, the overall heat is about two orders of magnitude smaller in comparison with the decay heat after 2 years. With half-lives of about 30 years, the influence of C137 and Sr90 decreases expectedly strongly towards 300 years and several actinides start to dominate the heat generation.

Table 4: Heat generated by the most dominant nuclides (98.8% of the generated heat) 40 years after reactor shut down for a representative 55 MWd /kg_{HM} burnup.

Z	nuclide	half time	decay heat (W)
56	Ba137m	2.552 m	4.997e+3
39	Y90	2.67083 d	4.714e+3
94	Pu238	87.6984 y	3.616e+3
95	Am241	432.804 y	3.385e+3
96	Cm244	18.0004 y	1.544e+3
55	Cs137	30.0406 y	1.509e+3
38	Sr90	28.7898 y	878.1
94	Pu240	6563.04 y	426.2
94	Pu239	24113.5 y	217.4
63	Eu154	8.59302 y	145.4

2.c Long-term repositories

At periods of about 40 to 50 years, the quantity of SNF that can be disposed together depends on its heat load and on the geological conditions. Salt rocks can conduct heat better than clay. However, for the safety of a waste disposal, the mobility of potential radioactive materials within the hosting rock becomes one of the crucial criteria for the loading scheme within the disposal casks, with the aim of controlled heat removal and radioactive dose in the storage gallery. Those issues are discussed in the next section.

As far as the nuclear data are concerned, the 4 tables highlight the main nuclides that should be carefully looked at to assure that the decay heat will be removed safely in wet as well as dry temporary storage and final disposal sites. Evidently, some of the nuclides are connected to chain processes (in particular Cm244), so that all nuclides involved in such chains should be in principle re-examined.

3. Nuclear needs for spent fuel in disposal site

The size and material composition of the HLW hosting casks is still an open issue. *At present, there are different criteria for the size and material composition of the hosting casks for high-level waste*. Different regulations in different countries would most probably change the final demands for each specific disposal site.

For example, in the planned final disposal in granite rocks in Finland and Sweden, a copper cover surrounding the casks is planned to avoid corrosion [4]. Another existing option is the Pollux cask, discussed within the German waste disposal program for final disposal in rock salt

[5]. In both cases, it is quite clear that iron would be a major element in such spent fuel casks. Therefore, iron nuclear data should be carefully checked, up to about 1 MeV.

In general, three types of host rocks seem to be suitable to host the nuclear waste. As mentioned before, different countries consider different solutions, in accordance with their available geological conditions. In the northern countries, as mentioned above, the host rock is mainly granite. Yet, salt rocks and clay rocks might have advantages for other countries. The advantages and disadvantages of each rock as far as their applicability to host radioactive waste can be found in [6]. For example, clay rocks have the advantage of immense sorption and very low solubility. A disadvantage is the lower heat conductivity in comparison with salt rocks. The storage capacity of clay rock is thereafter lower in comparison with salt rock. On the other hand, the small sorption and high solubility of the salt rocks introduce also complex challenges.

The nuclear data needs stemming from the above paragraphs on salt and clay rocks concern not only iron, but also the main nuclides within the rocks. For salt rocks one concentrates mainly on sodium Na23 and chlorine: Cl35, 36 and 37. Where Cl36 is a beta emitter with very long half-life time and can be generated by neutron absorption of Cl35.

The clay rocks are usually covered by concrete to gain stability of the gallery and here the list is quite long. Beside oxygen one has to look at different isotopes of silicon (Si28,29,30) and Ca. Further impurities are iron Fe, magnesium Mg and potassium K.

Preliminary studies, which are affected by the cross sections of the above-mentioned rocks, aimed to predict the neutron and gamma dose within the gallery in which the (neutron-emitter bearing) spent fuel is stored.

The scattering of neutron from the wall galleries to the gallery inner space increase the dose and reduces the allowed time for workers to be in the gallery. The issue of the secondary energy distribution, including energy and scattering angle is hence important. In particular, large angle scattering (back scattering) will reduce significantly the probability for neutrons to be absorbed within the wall. Further the larger the energy loss due to the scattering process the smaller will be the equivalent dose of the scattered neutrons.

Figure 1 and Figure 2 show the predicted (normalized) back scattering of neutrons from a salt and concrete wall respectively. The specific initial neutron energy spectrum emerging from the spent fuel container is shown in Figure 3. The collision bins show how many collisions were performed within the examined walls before the neutron was back scattered to the environment. Both figures demonstrate that there are cases of multiple scattering beyond 10, which emphasizes the importance of using a correct scattering formalism to account for the gallery wall, and in particular the angular distribution of the scattered neutrons. An additional important feature, which depends directly on the scattering cross sections of the nuclides involved, is the moderation rate. The appearance of considerable light elements within the concrete, such as oxygen, softens largely the neutron spectrum, and therefore the energy dependent neutron dose is reduced. Such aspects are of highly relevance for the safety of workers and the maintenance regulations within the gallery as discussed in [7].

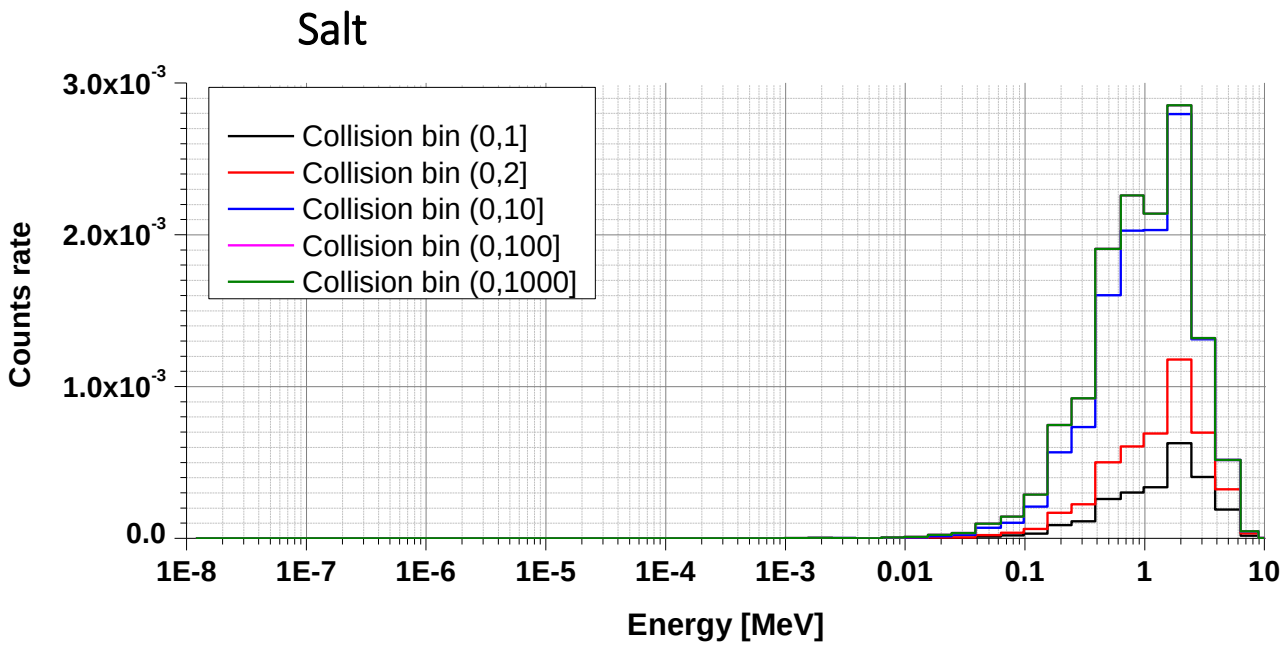

Figure 1: Energy dependent count rates of back scattered neutron from a 30 cm thick salt plate. The energy spectrum of the incident neutrons is taken from [17] and is plotted in figure 3.

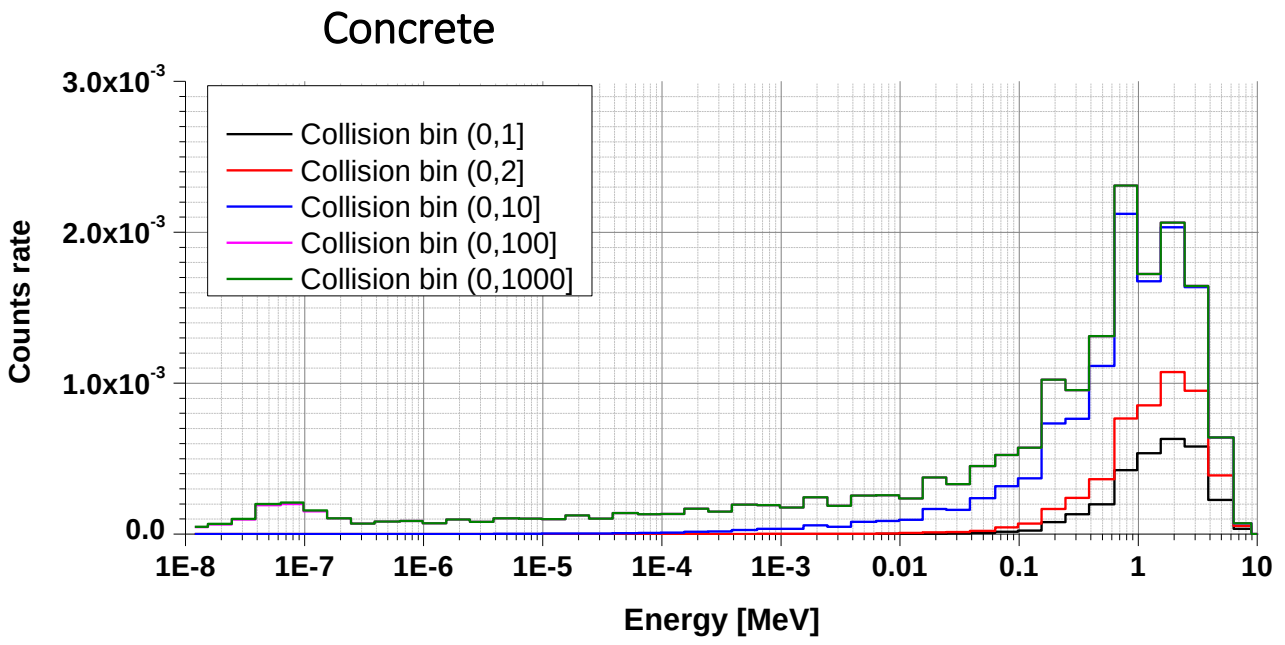

Figure2: Energy dependent count rates of back scattered neutron from a 30 cm thick concrete plate.

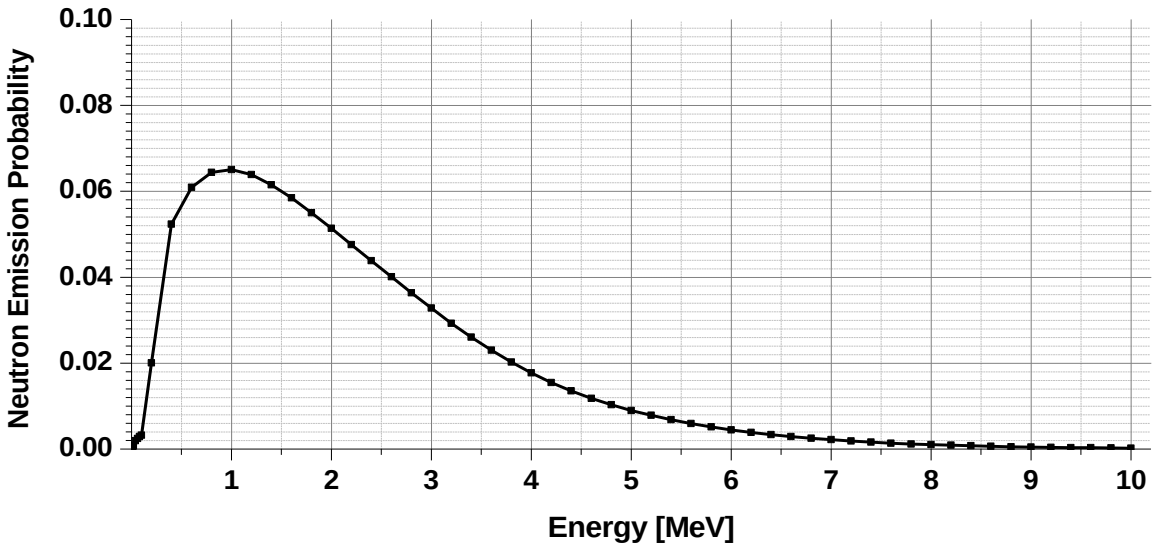

Figure 3: Energy dependent neutron emission probability from a loaded PWR spent fuel POLLUX 10 cask taken from [17].

A schematic yet realistic view of a gallery in which the casks could be deposited is shown in Figure 4. The white points in the figure are neutron and gamma detectors, which are being simulated by means of specific "Tallys" in MC codes, like MCNP. The reflective surrounding walls of the gallery emphasize the importance of the adequate angular distribution estimation of the neutrons on the nuclides making up the material composition of the walls. This type of gallery allows for more flexibility as the casks can be transported or placed close to each other if the radiation conditions have improved after a sufficient decay period. Further on, as one is concerned about the radiation field and corresponding dose within the gallery, the absorption rate of the nuclides and in particular, their emission spectrum, is of high interest.

As far as the POLLUX 10 type cask itself is concerned, the nuclear data of polyethylene embedded as tubes within the POLLUX cask should be looked at. Polyethylene absorbs and moderate neutrons, and thus reduces considerably the equivalent dose.

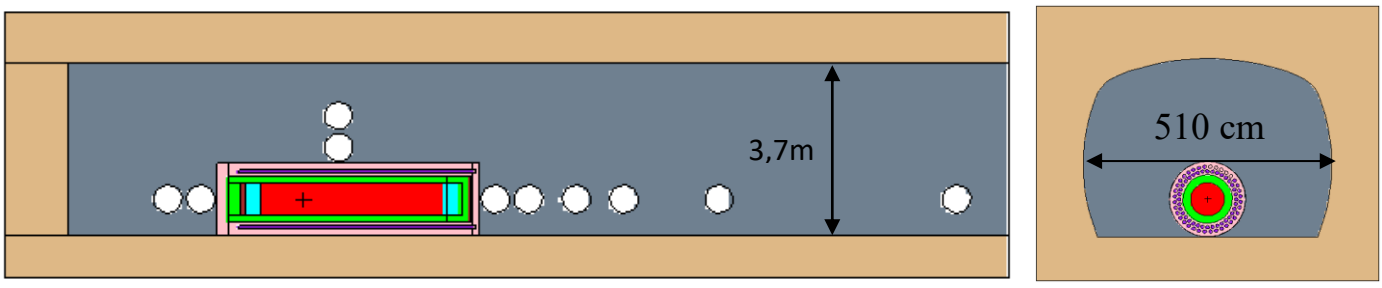

Figure 4: A longitudinal and a transverse cross section of a 15 m long gallery with a POLLUX 10 type cask. The transverse cut shows the polyethylene rods within the steel cladding of the casks. The white dots represent simulated neutron detectors for dose exposure.

The spectrum of the emitted neutrons from the casks expands over KeV and MeV energy range. Consequently, the investigation of the nuclear data, in particular for iron, should be extended to this energy domain. Further, in this KeV energy range, there are several options for the neutron scattering and different models show different results as was shown in [8]. For example, the iron scattering cross section is being often treated by the OMP (Optical model Potential). At the same energy range, of about 25 KeV, the MCNP [9] solution is using as default the "classical" momentum and energy dependent angular scattering kernel, calling for the basic Newton rules which at their very basics, differs completely from the quantum mechanics approach of OPM. On the other hand, in the ENDF-VIII [10] version another option is introduced based on the so called Blatt & Biedenharn Method [11],[12]. This option, which uses adapted orbital moments, based correction factors could be also adequate for particular energy regimes. Other relevant energy regimes could be fitted via the DBRC model [13] which is certainly better (at least as far as the physical conditions are concerned) than the 0 K approach mentioned above. Obviously, those scattering kernels should be further investigated. Here for a new dedicated transmission experiment at different temperatures up to 900 K is planned for the end of 2022 at the GELINA facility at GEEL [18].

Moreover, preliminary results show that the temperature impact on the scattering distribution is not negligible. In the lower KeV energy range, the temperature effect on the cross section is clearly visible. For example, the temperature impact for an incident neutron of 7.9 KeV (at the lower left wing of the first resonance of Cl-37) at 600K, interacting with the salt rock material Cl-37 is shown in Figure 5. The angular distribution domain for the scattered neutrons were divided into 20 equal cosine bins from -1 to 1. At 0 K it is well known that one should get equal distribution over the relevant energy range $E \to \alpha E$

Where:

$$\alpha = \left(\frac{A-1}{A+1} \right)^2$$

and A is the mass ratio between the nuclide and the neutron.

Figure 5 shows, however, that the real angular distribution is considerably different. The expected rectangular form of each bin, which corresponds to 0 K, is not valid. In particular, one can see an enhanced forward scattering, which reaches 13.3% between the 180° backwards scattering (black cosine bin -1 to -0.9) and the full forwards scattering 0° (orange cosine bin 0.9 to 1). In view of the multiple scattering that were shown in Figure 1 and 2 and in view of the dose rate estimation within the host gallery, one is inevitably lead to consider the temperature dependent scattering data in accordance with its numerical treatment in Monte Carlo (MC) solvers. Strictly speaking different temperatures means different scattering patterns leading to different does rates within the gallery.

20 Cosine bins:

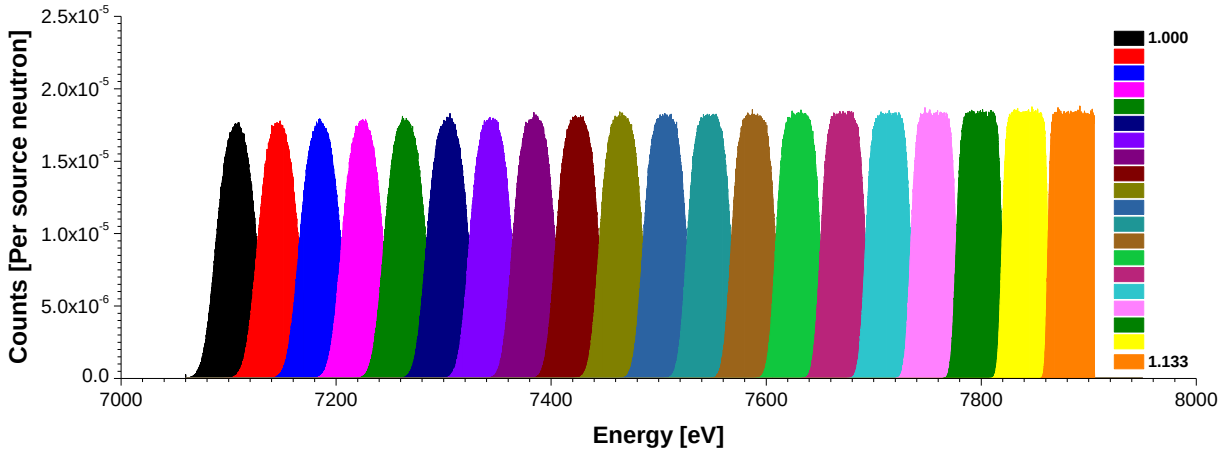

Figure 5: Angular distribution for an incident neutron of 7.905 KeV interacting with Cl37 at 600K. The first black bin -out of 20- is normalized to 1, so that the last orange cosine bin, between 0.9 to 1, has a scattering probability of 1.133 compared to the black cosine bin from -1 to -0.9.

The temperature effect as well as the geometrical effects that were handle in this section are summarized in Figure 6 for different materials with relevance with respect to the disposal site. Four types of gallery walls are introduced. The polyethylene (PET), which is actually presented within the cask and the air, are evidently two theoretical cases, defining the margin for the actual neutron counts, which one experiences at 1 m distance from the cask (white point in Figure 6). The PET and the concrete wall exhibit a considerable attenuation of the hard neutron spectrum (hence the dose) due to enhanced moderation by hydrogen and oxygen respectively. The reduced values in air show the importance of the absorption and scattering processes within the hosting rock. Evidently, for the air simulation, a neutron can be detected only once, when it arrives directly from the cask and cannot return by back scattering processes, like in the other cases, from the gallery walls. This means that the salt rocks contribute the most to the dose rate not only through the enhanced back scattered neutrons but although through the lack of moderation which increases the equivalent dose rate as fast neutrons have a larger weight dose factor (3-4 times more than thermal neutrons).

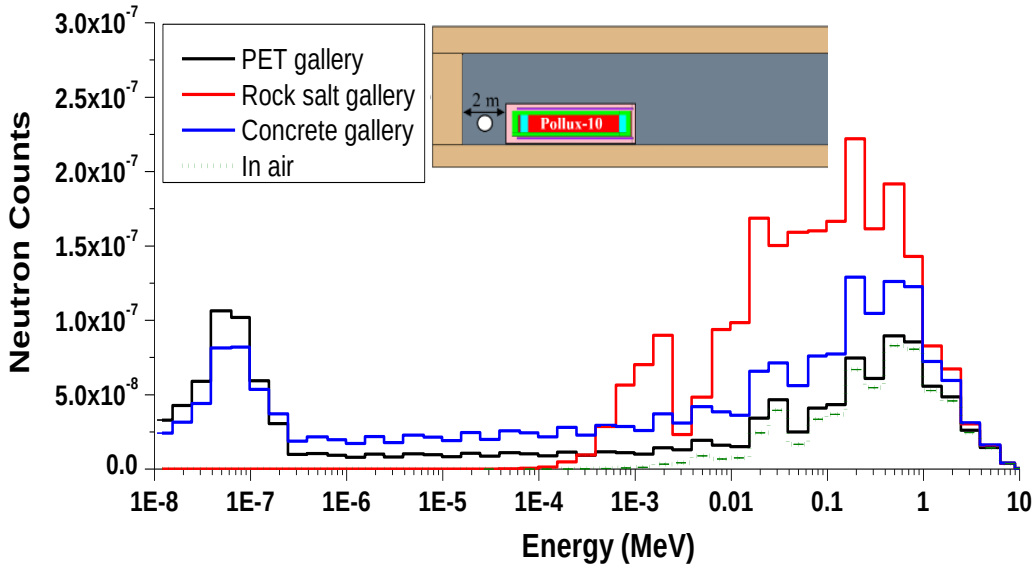

Figure 6: Neutron counting rates at a detector at 1 m distance along the axis of the POLLUX type cylindrical cask. Four different material layers were depicted at postulated 600 K hot casks.

4. Mobility and induced corrosion of radioactive nuclides

Mobile radionuclides present in SNF

The composition of SNF differs significantly from the starting material UO2 or MOX [14,15]. During reactor power generation, two substantial reactions take place, which affect the chemical behaviour and homogeneity of the initial fuel: Fission products formation and in addition, transuranic isotopes are generated because of neutron capture and subsequent (mostly) α *or* β^- decay starting from U238. The ultimate composition of the SNF depends on various factors e.g. type of fuel, initial enrichment, burn-up and linear power rating of the fuel during reactor operation. Additionally, and in view of the nuclear data needs, activation products produced due to neutron capture by trace impurities within the fuel and cladding are distributed in various parts of the fuel rods, e.g. Cl36 and C14 in the gap region, cracks and in the cladding. Further mobile radionuclides, which include fission gases and fission products (Tc-99, Kr, I-129, Xe, Cs-135/137 and Se-79), migrate during reactor operation to the pellet-cladding gap, grain boundaries and to cracks in the fuel [14-16]. In that sense, determining the specific actinides, activation and fission products quantities in different physicochemical states, allows for a better prediction of their chemical behavior (integrity of the fuel rods) and transport mobility (biological environmental risk). Consequently those latter issues are of major importance for the optimized management of SNF for interim storage, conditioning and final disposal.

Behavior of SNF under dry interim storage

After discharge of SNF from a nuclear power reactor and subsequent cooling in a spent fuel pool for several years, the fuel assemblies are possibly placed in dry cask storage for some time before transfer for final disposal in a deep geological repository.

Integrity of the irradiated Zircaloy cladding after 50 to 100 years of dry interim storage is essential e.g. for conditioning of the fuel assemblies for final disposal. However, the cladding is affected during reactor operation by various processes such as elongation of the fuel rods due to creep behaviour and oxidation with the water coolant, causing a reduction of the Zircaloy wall thickness, respectively. Further, dissolution and precipitation of hydrogen within the Zr-alloy matrix, during reactor operation, possibly leads to hydrogen embrittlement and delayed hydride cracking of the cladding. Swelling of the fuel pellets during irradiation due to fission products build-up eventually leads to the contact of the fuel pellets with the cladding. The so-called pellet/cladding interaction (PCI) induces tensile stress on the cladding, especially during power ramps. Moreover, precipitates of volatile fission/activation products such as Cs-135/137, I-129, Rb, Te, and Cl-36 present on the inner surface of the cladding or at the fuel-cladding interface possibly exhibit corrosive properties and thus affect the cladding integrity. Another phenomenon affecting the physical properties of the cladding during interim dry storage is the irradiation damage produced in the inner surface of the cladding by the alpha decays of the actinides present at the periphery of the pellet, especially when the burn-up at discharge is high.

Behavior of SNF in a deep geological repository

In safety assessments for disposal of spent nuclear fuel in a deep geological repository, water access, consecutive failure of canisters and loss of integrity of fuel cladding are considered in the long-term [14,15]. In this context, it is indispensable to evaluate the contribution of fission- and activation products as well as gases to the instant release fraction (IRF). This fraction consists of mobile, long-lived radionuclides with low retention tendency within the repository and contributes significantly to the calculated doses arising from release in a deep geological repository.

Upon contact of ground water with the SNF, a rapid release of radionuclides such as Cs-135/137, I-129, Cl-36, Se-79, Tc-99 and fission gases (Xe, Kr), amongst others, from these highly reactive sites is expected [14,15]. These radionuclides are easy soluble with low retention capabilities due to their mostly anionic character.

5. Conclusions

The nuclear data needs for spent fuel were discussed in this SANDA Deliverable 5.4 relative to three different aspects of SNF disposal.

The decay heat issue, which involves improved data on absorption and fission of actinides on one hand and the fission yields of the fissionable materials on the other. The decay heat was shown to have an impact on short-term handling of the fuel, namely the duration of the necessary forced flow conditions and the meaning of natural convection in wet storage, as was unfortunately shown in the melting of fuel rods in the wet storage in Fukushima. In midterm handling, the decay heat is of the key issues in designing the type of casks in temporary dry storage. For the final disposal within dedicated underground galleries, the decay heat in combination with the heat conductivity of the hosting rocks influence directly the quantity and volume of the allowable spent fuel to be stored.

The further need of investigation was emphasized this year 2021 by the opening of the OECD sub group investigation community called “Decay heat” within the WPNCS –Work Package Nuclear Criticality Safety.

The mobility of certain radioactive nuclides and their particular biological hazard lead to the need for further investigations of nuclides that usually are not of big importance in reactor physics. To this category belong, among others C-14, Se79, Cs-135/137, I-129, Cl-36 and Tc-99, [16].

The final disposal site requirements in terms of nuclear data needs was elaborated. Besides iron, the material compositions of the hosting rocks were shown to be crucial for the dose levels within the galleries. It was shown that besides the absorption rates, neutron scattering effects on gallery walls and in particular the angular distribution for neutrons within the keV range, are of importance. Further, the impact of the temperature on those processes must be looked at. Note that a new project starting in 2022 at IRMM, Geel will evaluate the current treatment of temperature dependent cross sections.

It is evident that the nuclide list described in this study contains only the most known important elements based on the current options considered for nuclear waste disposal. This list could be to some extent extended or changed according to the specific regulation in each country.

The issue of the uncertainties of the nuclear data mentioned in this deliverable is dealt separately in general manner for spent fuel applications in task 5.2.

6. References

1. Lamarsh J., Baratta J., “Introduction to Nuclear Engineering”, Pearson Publishers.
2. Nucleonica GmbH, 2017, Nucleonica Nuclear Science Portal (www.nucleonica.com), Version 3.0.269, Karlsruhe
3. Jannson P. , Bengtsson M. et al. “Blind Benchmark Exercise for Spent Nuclear Fuel Decay Heat”. To be published 2022
4. Hedin et al. (2007) NEA-RWM report, NEA No. 6362, Nuclear Energy Agency, Paris, pp 45-56
5. Janberg, K., & Spilker, H. (1998)."Status of the development of final disposal casks and prospects in Germany" Nuclear Technology, Vol. 121, pp 136-147
6. Federal German Institute for Geosciences and raw materials report (2007) (in German) „Untersuchung und Bewertung von Regionen mit potenziell geeigneten Wirtsgesteinsformationen“ Hannover / Berlin.
7. H. Suarez „Individual dosimetry in disposal facilities for high-level nuclear waste”, KITopen, DOI: 10.5445/IR/1000084032
8. R. Dagan, A. Konobejeev, “Extended Validity of the Energy Dependent Scattering Kernel within the Boltzmann Transport Equation”. DOI: 10.1080/23324309.2020.1836497, Journal of Computational and Theoretical Transport 2020.
9. MCNP6 Users Manual - Code Version 6.1.1beta, LA-CP-14-00745 (June 2014).
10. D.A.Brown, M.B.Chadwick et al. ”ENDF/B-VIII.0: The 8th Major Release of the Nuclear Reaction Data Library with CIELO-project Cross Sections, New Standards and Thermal Scattering Data” Nuclear data sheets vol. 148 pp 1-142
11. Blatt J. M. , L.C. Biedenharn, „The angular Distribution of Scattering and Reaction Cross sections“, Rev. of modern Physics, Vol. 14 Oct. 1952.
12. F. Gunsing “Resonances in neutron-induced reactions “ Eur. Phys. J. Plus (2018) 133: 440.
13. R. Dagan, B. Becker, Y. Danon, M. Rapp, D. Barry, G. Lohnert, ”Modelling a resonance dependent angular distribution via DBRC in Monte Carlo codes”, International Conference on Nuclear Data for Science and Technology, Jeju island, KOREA 2010. J. Korean Phys. Soc. 59: pp.983-986, 2011.
14. J. Bruno, R.C. Ewing, “Spent Nuclear Fuel”, Elements, Vol. 2, 2006, 343-349.
15. R.C. Ewing, “Long-term storage of spent nuclear fuel”, Vol. 14, 2015, 252-257.
16. B. Grambow, “Mobile fission and activation products in nuclear waste disposal”, Journal of Contaminant Hydrology, Vol. 102, 2008, 180-186.
17. Saurí Suárez, H., et al. "Neutron flux measurements on a mock-up of a storage cask for high-level nuclear waste using 2.5 MeV neutrons." Journal of Radiological Protection 38.3 (2018) 881-891.
18. P. Schillebeeckx , private communication, GELINA facility Geel , 2020.