Technical Report
TR-19-14
February 2019

Analysis of radiation damage in the KBS-3 canister materials

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ISSN 1404-0344
SKB TR-19-14
ID 1708927
February 2019

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Abstract

Here are presented calculations of neutron and gamma radiation damage rates and total damage that spent fuel assemblies give rise to in the materials of the KBS-3 design (copper canister and cast iron insert). Previous calculations by Guinan (2001) were repeated using recently developed techniques in order to check the validity of his conclusions. The program SPECTRA-PKA and the data library TENDL processed by the NJOY code were used to calculate the neutron radiation damage on Fe and Cu. Results of this work fit with the reference very well. We also evaluated the total amount of hydrogen and helium produced and copper diffusivity over 10^5 years. Based on our results, we conclude that radiation effects should not significantly damage the canisters after 10^5 years under the considerations detailed in this report.

Sammanfattning

Här presenteras beräkningar av strålskadedosrater från neutron- och gammastrålning som använt kärnbränsle kommer ge upphov till i materialen i KBS-3 designen (kopparkapseln och segjärns-insatsen). Tidigare beräkningar av Guinan (2001) upprepades med hjälp av nyligen utvecklade metoder för att jämföra med hans slutledningar. Programmet SPECTRA-PKA och tvärsnittsbiblioteket TENDL användes för att beräkna neutronstrålningens skadedos i järn och koppar. Resultaten av detta arbete stämmer väl överens med referensen. Vi utvärderade även mängden väte och helium som byggs upp genom transmutation samt kopparns diffusivitet över hundra tusen år. Vi slutleder utifrån våra beräkningar att strålningen inte kommer skada kapselns material nämnvärt under hundra tusen år, utifrån de antaganden som gjorts i denna rapport.

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1 Introduction

According to the KBS-3 method, spent nuclear fuel is emplaced in copper canisters surrounded by protective bentonite clay and deposited at about 500 m depth in granitic rock. The role of the canisters is to contain the spent fuel for as long as required in order to protect man and the environment from harmful effects of ionizing radiation from the waste. The safety assessment of a spent fuel repository encompasses typically one million years after closure of the repository.

The atoms in a crystalline solid are arranged in a three dimensional periodic structure which minimizes the free energy of the system. This structure is never perfect but defects such as vacancies, grain boundaries and dislocations are always present. These are important as they contribute to the ductility of the material and are responsible for its ability to deform plastically under stress. However, when exposed to ionizing radiation crystalline damage may occur, potentially leading to redistribution of atoms as well as rejection of solutes from the crystal. In this case the chemical composition of the system may be altered and the material risk suffering from premature aging and mechanical failure. The goal of this work is to evaluate the radiation damage effects on the copper shell on the KBS-3 canisters from neutron and γ-irradiation.

Radiation damage occurs in a crystal if the transmitted energy from the energetic particles is sufficient to create stable defect pairs (Frenkel pairs) in the lattice. Historically, the Norgett-Robinson-Torrens analytical calculation method for the number of displacements per atom (dpa) in a lattice (NRT-dpa) (Norgett et al. 1975) has been the standard evaluation method for this purpose during past decades, beside the Stopping Range of Ions in Matter (SRIM) code package. More modern approaches use semi-empirical interatomic potentials and classic molecular dynamics (MD) simulations. Recent progress has opened up for fully quantum mechanical calculations of the primary damage state and radiation damage parameters (Olsson et al. 2016).

The damage evolution in the KBS-3 design has been evaluated with the conclusion that no significant damage should occur (Guinan 2001). However, some doubts regarding these results were raised in a theoretical study that radiation enhanced solute clustering effects in the cast iron could lead to degradation of the mechanical properties (Brissonneau and Bocquet 2003, Brissonneau et al. 2004). The KBS-3 design parameters took this into account and in a more recent study using both a theoretical and experimental investigation of the cast iron evolution, lead to the conclusion that the current design parameters should be safe, with respect to Cu-induced embrittlement in the cast iron matrix (Olsson et al. 2013, Chang 2015). Now we aim to re-evaluate the radiation damage effect in the copper shell as well, and to study possible transmutation and generation of light elements that could potentially lead to issues in long term operation.

2 Methodology

In order to evaluate the total damage, we need to calculate the damage rate first. Here we establish the theoretical framework for displacement damage. First the damage rate equation is presented as the equation governing the impact of radiation field on the material. After this its various components are discussed together with equations relevant in the context of the final repository. The difference between gamma induced damage and neutron induced damage will be discussed here.

2.1 Displacement damage

When the incident radiation interacts with atoms in a crystal, all or some of the energy may be transferred. If the energy is sufficient, an atom may be scattered from its position in the lattice and propagate through the material. As it moves in the material it will interact with the surrounding atoms and either recombine back to its original position, stabilize as a Frenkel pair (a vacancy and a self-interstitial) or generate a displacement cascade if the energy is high enough. The total number of displaced atoms in the metal due to an incident projectile is given by the damage rate equation (Was 2007), Equation 2-1.

$$R_d = N \int_{\check{E}}^{\hat{E}} \phi\,(E_i)\sigma_D(E_i)dE_i \tag{2-1}$$

where N is the atom number density, $\phi(E_i)$ is the energy-dependent particle flux and $\sigma_D(E_i)$ is the displacement cross section, see subsection 2.1.1. The damage rate is given in dpa/(unit time × unit volume).

2.1.1 Displacement cross section

The displacement cross section, given by Equation 2-2, is the probability that a nucleus will be displaced from its position in the lattice by an incident particle (Was 2007).

$$\sigma_D(E_i) = \int_{\check{T}}^{\hat{T}} \sigma_S\,(E_i, T)\nu(T)dT \tag{2-2}$$

where $\sigma_s(E_i,T)$ is the scattering cross section – the probability that an energy will impart a recoil energy T to the struck atom – and $\nu(T)$ is the number of displaced atoms as a function of transferred energy, discussed in subsection 2.1.2.

2.1.2 Number of displaced atoms per impact

The minimum energy needed to displace an atom from its place in the crystal lattice is known as the threshold displacement energy, E_d. If an atom is displaced from its lattice site, it receives a kinetic energy given by $E_K = T–E_d$, where T is the energy transferred in the collision. If the kinetic energy is high enough, the primary knock-on atom (PKA) will move in the crystal lattice and when doing this it may impact the surrounding atoms and a displacement cascade may follow. The number of displaced atoms, $\nu(T)$, as a function of transferred energy can be modelled by the Kinchin-Pease model, presented in Equation 2-3 (Was 2007).

$$\nu(T) = \begin{Bmatrix} 0 & T < E_d \\ 1 & E_d < T < 2E_d \\ \frac{T}{2E_d} & 2E_d < T < E_c \\ \frac{E_c}{2E_d} & T \geq E_C \end{Bmatrix} \tag{2-3}$$

where T is the transmitted energy, E_d is the average threshold displacement energy and E_c is the cut-off for when electronic stopping starts to dominate. However, this model is not accurate for the low (near threshold) nor high energy ranges (far above threshold). For high energies the NRT model (Norgett et al. 1975) has been seen as more appropriate but the recently developed arc-dpa model is clearly more realistic (Nordlund et al. 2018). However, here the dominating damage processes are in the lower and intermediate energy ranges and an in-depth discussion regarding the high-energy recoil range will therefore not be pursued. For the lower energy range, it has been shown that the energy required to displace an atom is not a clear threshold, but in fact depends on the angle at which the atom is scattered. A classical molecular dynamics simulation of the energy required to displace a copper atom as a function of direction is presented in Figure 2-1 (King et al. 1983). As seen in this figure, sub-threshold effects are important in order to properly capture the displacement behavior.

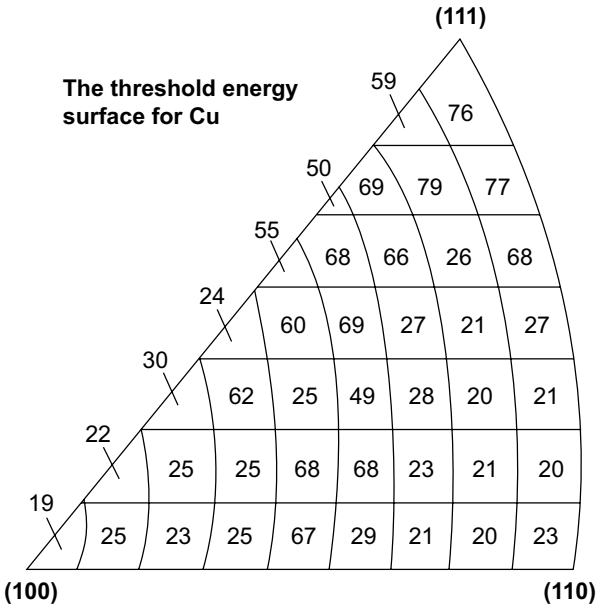

Figure 2-1. *Threshold displacement energy in eV as a function of direction in copper. From King et al. (1983).*

2.2 Photon-electron interactions

For the gamma induced damage, energy can be transferred indirectly from a photon to the copper ions through pair production, photoelectric absorption, or Compton scattering of electrons, which may in turn interact with the ions. The energy of the γ-radiation considered is not high enough for pair-production to be possible, thus only direct photon-electron interactions are considered in this work.

2.2.1 Compton scattering

In the case of Compton scattering, the hard-sphere approximation gives the energy transferred from the photons to the electrons as a function of the photon's scattering angle. The result is given by Equation 2-4.

$$T_{Compton} = \frac{E_\gamma^2(1-\cos\theta)}{E_\gamma(1-\cos\theta)+m_e c^2} \tag{2-4}$$

The maximum transferred energy, $\hat{T}$, is given in the case of total backscattering of the photon, that is $\theta = \pi$. This is given by Equation 2-5.

$$\hat{T}_{Compton} = \frac{2E_\gamma^2}{2E_\gamma+m_e c^2} \tag{2-5}$$

In the case of the 661 keV photons considered in this work, the maximum energy transferred to an electron through Compton scattering is 477 keV.

2.2.2 Photo-electric absorption

In the case of photo-electric absorption the incoming photon transfers all of its energy to an electron, which is liberated. The electron receives a kinetic energy equal to the incident photon energy (minus the work function, which is a few eV for transition metals and can thus be ignored). However, as was shown in Toijer (2014) the cross section for photo-electric scattering is much smaller than that of Compton scattering for 661 keV gammas, so we can ignore this component.

2.2.3 Electron-nucleus energy transfer

In the event of elastic scattering of an electron from a lattice nucleus, the energy transferred from an electron to a nucleus is given by the hard-sphere approximation as a function of the electron's scattering angle, ϕ (Was 2007):

$$T = \frac{2E_i}{Mc^2}(E_i + 2m_e c^2)\sin^2\left(\frac{\phi}{2}\right) \quad (2\text{-}6)$$

where where M is the mass of the ion, E_i is the electron's energy, that is $T_{Compton}$ in Equation 2-4 in the case of Compton scattering and E_γ in the case of photoelectric absorption. In the case of Compton scattering, this results in an energy transferred to the nucleus as a function of both photon and electron scattering angles, given by Equation 2-7.

$$T = \frac{2}{Mc^2}\left(\frac{E_\gamma^2(1-\cos\theta)}{E_\gamma(1-\cos\theta+m_e c^2)}\right)\left(\frac{E_\gamma^2(1-\cos\theta)}{E_\gamma(1-\cos(\theta))+m_e c^2} + 2m_e c^2\right)\sin^2\left(\frac{\phi}{2}\right) \quad (2\text{-}7)$$

where θ is the γ-scattering angle and ϕ is the electron scattering angle. In total, the maximum energy transferred from a 661 keV gamma photon to a copper atom via Compton scattering is 24 eV and for iron it is 27 eV. Both these values lie below the two metals' respective average threshold displacement energies and as such the canonical damage models predict no damage at all. However, taking the sub-threshold effects into account, which was not properly done in previous studies, there will be some damage created in a fraction of the scattering solid angle. The transferred energy is also clearly low enough to only create single Frenkel pairs.

2.3 Neutron-ion interactions

For the interaction of energetic neutrons with ions, the same treatment as in Section 2.1 is valid. However, the neutrons have smaller cross sections and longer mean-free paths but typically depose damage in a much more localized manner. Neutrons can interact with ions (or nuclei rather) in a few distinct ways, namely elastic scattering, inelastic scattering, or through nuclear reactions, such as (n,p), (n,2n) or (n,α) reactions. For elastic scattering, hard-sphere kinematics (Was 2007) give a maximum energy transfer T to an ion as

$$T = \frac{2AE_n}{(A+1)^2}(1 - cos\,\phi) \quad (2\text{-}8)$$

where A is the atomic mass number of the PKA, E_n is the incident energy of the neutron and ϕ is the scattering angle. Thus, a 1 MeV neutron will on average transmit about 35 keV to the PKA in iron, and 30 keV to that in copper, generally three orders of magnitude higher energy than the damage threshold. This type of high-energy PKA will always generate displacement cascades instead of single Frenkel pairs, as opposed to the case for gamma (or indirectly, electron) irradiation. The treatment of neutron induced damage is thus more complicated and less easy to deal with analytically. Here, we apply a method and code (SPECTRA-PKA) (Gilbert 2018) developed recently that is capable of calculating PKA spectra, damage levels and nuclear reactions for neutron-ion interactions.

Based on a nuclear database, for a given nuclear reaction on a target (or parent) nuclide, the recoil energy distribution for the daughter nuclide is readily available for neutron interactions. The code can be used with any nuclear cross section library. Here we have used SPECTRA-PKA with the TENDL-2017 database (Koning et al. 2017) processed by the NJOY code (MacFarlane 2012, Rochman 2017). A major reason why we apply the SPECTRA-PKA code instead of SPECTER, as was used by Guinan (2001) is the possibility to study all possible reaction channels concurrently and consistently and to be able to use the most modern nuclear data library (TENDL). The choice of using a different code and cross section library also allow us to determine if the choice of code matters on the damage calculations. SPECTRA-PKA applies the NRT model for damage calculations (Norgett et al. 1975, Gilbert and Sublet 2018) and thus the dpa rate for the given $i \rightarrow j$ reaction channel R_{ij} is:

$$R_{ij} = \frac{0.8}{2E_d}\sum_n \phi_n \sum_k \sigma_{kn}^{ij} T_j^k, \quad (2\text{-}9)$$

where E_d is the threshold displacement energy; ϕ_n is the neutron flux ($cm^{-2}s^{-1}$) at neutron energy E_n; σ_{kn}^{ij} is the cross section in barns for a recoil in energy bin k for an incident neutron at energy E_n for reaction $i \rightarrow j$; T_j^k is the damage energy for daughter j at energy bin k. The total dpa rate of a material can be obtained from Equation 2-9 by summing over j possible reactions and then over i different reaction targets with a relative concentration weighting. In this way, both damage rate and transmutation rates can be calculated.

3 Results and discussion

Damage rate calculations have been performed for gamma and neutron irradiation effects in copper, and in some cases in iron.

The canister configuration for the PWR fuel assembly used by Guinan (2001) is shown in Figure 3-1.

3.1 Decay of spent fuel

The relative impact of the various radioactive isotopes of nuclear fuel have been previously examined. In a work by Toijer (2014), the γ-activity per metric tonne of uranium after 15 years of interim storage was used, weighted with the energy of the emitted radiation, for BWR fuel at 55 MWdays/kg uranium. The results are presented in Figure 3-2.

As seen in Figure 3-2, ^{137}Cs is by far the most relevant species in terms of energy and activity in the first few centuries. The decay chain from ^{90}Sr is of secondary long-term importance. These results were similar in the case of the PWR fuel. ^{137}Cs decays through beta-emission to metastable ^{137m}Ba that in turn decays to stable ^{137}Ba through the emission of 661 keV γ-rays. This decay chain dominates the activity and the generated radiation damage and thus ^{137}Cs is the only isotope considered in the following.

In order to assess the direct effects on the copper from the γ-irradiation, the damage rate equation, Equation 2-1, should be solved. Since it has been established above that primarily electrons from photon-scattering can be responsible for damage in the material, the flux of electrons from photon interactions, and their subsequent impact on the copper atoms, will be calculated.

The neutron fluxes have been calculated previously by Håkansson (2000) and Guinan (2001). In Table 3-1 we show the neutron fluxes used by Guinan (2001) with which we compare first, in order to assess the choice of nuclear data library and method of calculation.

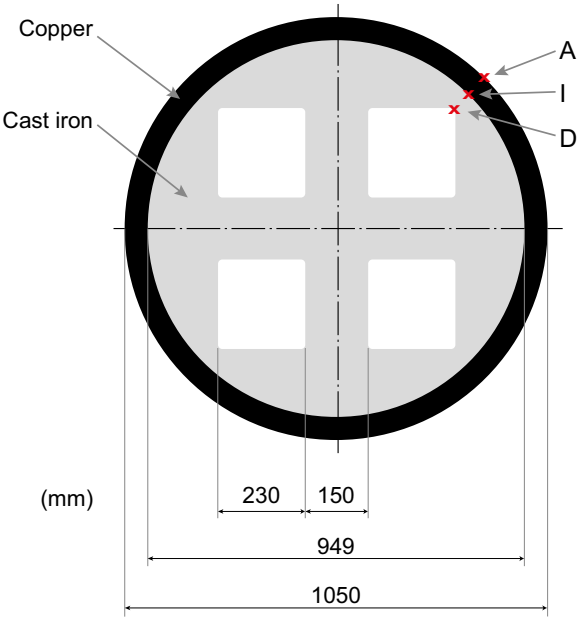

Figure 3-1. *The canister configuration for the PWR fuel assembly from Guinan (2001). Points A, I and D are henceforth used in the calculations.*

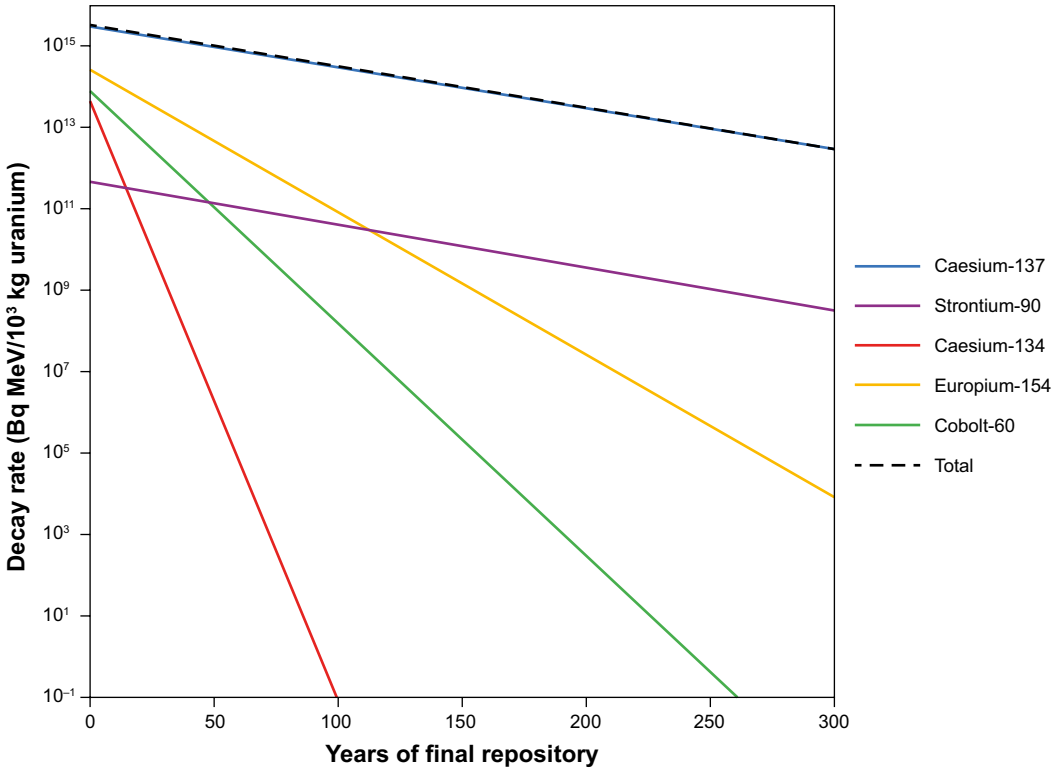

Figure 3-2. *γ-activity per metric tonne of uranium after 15 years of interim storage weighted with photon energy.*

Table 3-1. Neutron flux vs binned neutron energy at different positions of a PWR canister. Neutron fluxes are known at points A and D and Neutron flux at I were interpolated using neutron attenuation coefficients for both metals. See Table 1 and 2 in Guinan (2001).

Energy group (MeV)		PWR group flux, 3.5 % enrichment at 30 years (n/cm²s)		
Upper	Lower	At D	At I	At A
1.000E+01	6.065E+00	3.964E+02	2.110E+02	1.145E+02
6.065E+00	3.679E+00	1.801E+03	8.992E+02	4.728E+02
3.679E+00	2.231E+00	4.457E+03	2.244E+03	1.241E+03
2.231E+00	1.353E+00	6.182E+03	3.407E+03	1.986E+03
1.353E+00	8.210E–01	7.237E+03	4.644E+03	2.810E+03
8.210E–01	5.000E–01	1.144E+04	7.133E+03	4.251E+03
5.000E–01	1.110E–01	2.222E+04	1.249E+04	6.984E+03
1.110E–01	9.118E–03	9.944E+03	4.500E+03	1.766E+03
9.118E–03	5.530E–03	8.282E+02	2.904E+02	9.144E+01
5.530E–03	1.487E–04	6.519E+02	4.831E+02	2.333E+02
1.487E–04	1.597E–05	1.683E+02	1.071E+02	3.457E+01
1.597E–05	9.877E–06	1.272E+01	7.119E+00	3.547E+00
9.877E–06	4.000E–06	8.768E+00	5.150E+00	2.599E+00
4.000E–06	1.855E–06	2.723E+00	1.535E+00	7.286E–01
1.855E–06	1.097E–06	1.238E+00	8.059E–01	4.500E–01
1.097E–06	1.020E–06	8.488E–02	6.312E–02	4.258E–02
1.020E–06	6.250E–07	3.451E–01	2.369E–01	1.405E–01
6.250E–07	3.500E–07	1.936E–01	1.307E–01	7.738E–02
3.500E–07	2.800E–07	3.772E–02	2.762E–02	1.817E–02
2.800E–07	1.400E–07	5.621E–02	3.989E–02	2.478E–02
1.400E–07	5.800E–08	2.405E–02	1.831E–02	1.260E–02
5.800E–08	3.000E–08	5.056E–03	3.986E–03	2.877E–03
3.000E–08	1.000E–10	2.835E–03	2.102E–03	1.405E–03

3.2 Gamma photon flux

The flux of gamma photons in the case of BWR fuel (38 MWd after 30 years of interim storage) is given by Håkansson (2000). In the range of 511–800 keV, it is $\phi_0 = 6.353 \times 10^{12}$ photons/(m^{2}s) at the inner boundary of the copper. As the photons travel through the material, the intensity will be attenuated as described by the Beer-Lambert law. The flux of photons as a function of the distance from the copper-iron interface is given by Equation 3-1.

$$\phi_\gamma(x) = \phi_0 e^{-\mu x} \tag{3-1}$$

where μ is the linear attenuation coefficient, which is calculated to be 0.68 cm^{-1} for copper from the mass attenuation coefficient given by NIST. From this the penetration depth of the photons in copper is calculated to be $\delta = 1.46$ cm. The total linear density of photons deposited in the copper is given by Equation 3-2.

$$\chi_{tot} = \delta\, \phi_0 \left[1\text{-e}^{-\mu\delta}\right] = 5.86 \cdot 10^{10}\ (ms)^{-1} \tag{3-2}$$

3.3 Electron flux

Assuming that a total linear density of 5.86×10^{10} photons (ms)$^{-1}$ were deposited in the material, the electron flux from these interactions was calculated using the following equation

$$\phi_e = \sigma_s \chi_{tot} N_{Cu}\ (m^2 s)^{-1} \tag{3-3}$$

where σ_s is the γ-electron scattering cross section, different for the two interactions respectively, and N_{Cu} is the copper volume density, 8.5×10^{28} (atoms/m^3), assuming one conduction electron per atom. As seen in the work by Toijer (2014), the flux of photo-electrons is seven orders of magnitude lower than the flux of Compton electrons in the case of the iron part of the canisters. It is therefore considered a valid assumption that the importance of photo-electrons in the case of the copper is very low and these are omitted in the following calculations.

The energy differential cross section, calculated as in Toijer (2014), as a function of kinetic energy transferred to copper electrons for 661 keV γ-photons in copper is presented in Figure 3-3.

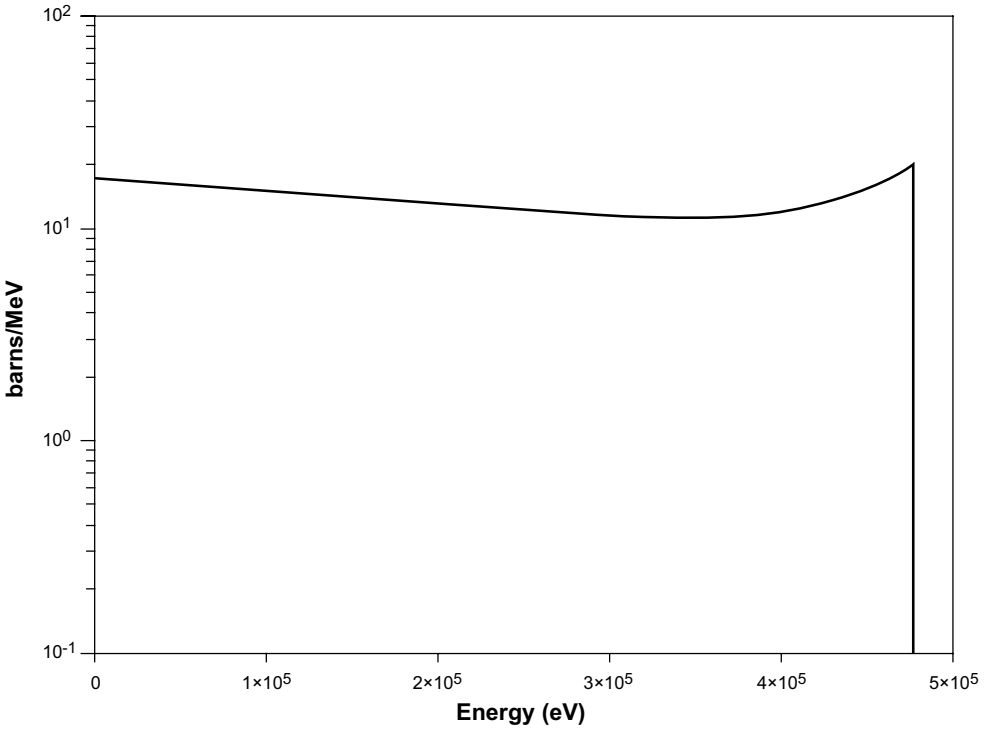

Figure 3-3. *Energy differential cross section as a function of kinetic energy transferred to the copper electron in the interaction with a 661 keV-γ-photon.*

The total Compton scattering cross section is given by Equation 3-4.

$$\sigma_C = \int_0^{\hat{T}} \left(\frac{d\sigma}{dE_K}\right) dE_K \tag{3-4}$$

From this the total flux of Compton electrons was calculated to

$\phi_{0,C} = 2.5 \cdot 10^8$ (m^{2}s)$^{-1}$.

Due to their electric field, the electrons produced from interactions with the γ-radiation can interact with the surrounding material and lose energy continuously as they propagate through the lattice. Thus, unlike the case of photons, the electron energy varies along its path. The change in energy as the electron propagates is known as the stopping power, S_p, of the material

$$S_p = \frac{dE}{dx} \tag{3-5}$$

The average electron energy as a function of distance traveled in the material is given by Equation 3-6.

$$E_e(x) = E_{e,0} - S_p x \tag{3-6}$$

As discussed above, the minimum energy transferred to a copper nucleus which can create a displacement is 19 eV. In order to transfer this amount of energy in the case of a head on collision, the electron energy needs to be at least 400 keV. The maximum amount of energy an electron can require through Compton scattering is 477 keV. The range of the 477 keV electrons before they are relaxed to 400 keV is calculated using Equation 3-6 to $R = 6.4 \times 10^{-5}$ m. This distance is insignificant with respect to the penetration depth of the γ-photons and it is thus assumed that all displacements occur within the photons' penetration depth.

3.4 Gamma damage rate

The total amount of displaced atoms from the electron flux given in Equation 3-3 was calculated based on the equations above. In the calculations a modified Kinchin-Pease model, given by Equation 3-7, was used to account for the displacements in the lower energy ranges.

$$\nu(T) = \begin{cases} 0 & T < E_{min} \\ 0.5 & E_{min} < T < E_d \\ 1 & E_d < T < 2E_d \\ \frac{T}{2E_d} & 2E_d < T < E_c \\ \frac{E_c}{2E_d} & T \geq E_C \end{cases} \tag{3-7}$$

where E_{min} = 19 eV and E_d = 30 eV for Cu, see Figure 2-1, are chosen to be consistent with the discussion by Was (2007).

From this the initial damage rate (after 30 years cooling) at the inner boundary of the copper capsule is calculated to 5.5×10^{-11} dpa/year. Following the change in activity due to the decay of ^{137}Cs, the damage rate will decrease with time. The total number of displacements per atom and time during the first 300 years of storage is plotted in Figure 3-4. The initial damage rate obtained by Guinan (2001), 1.0×10^{-10} dpa/year, is included for comparison.

From this the total gamma induced damage during the first 300 years of storage was calculated to $R_{tot} = 2.4 \times 10^{-9}$ dpa. Considering the small change with respect to earlier results by Guinan (2001), that is a factor of about 2 less damage rate, we reach the same conclusions here as well regarding the negligible effect of the gamma induced radiation damage over the full life span of the repository.

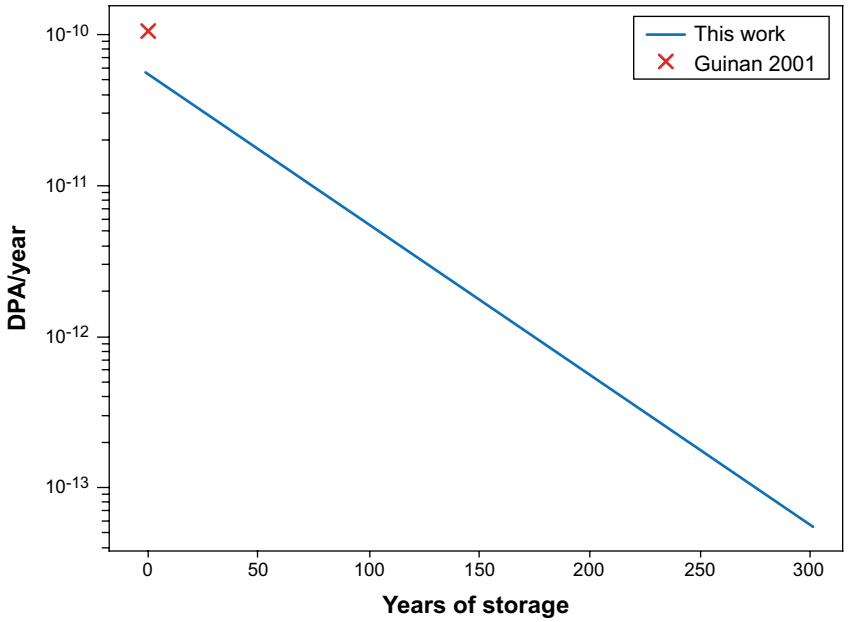

Figure 3-4. *Damage rate in the copper canisters during the first 300 years of final disposal. Results are compared with previous calculations performed by Guinan (2001).*

3.5 Neutron damage rate

As this work and Guinan (2001) used different codes and databases to calculate the dpa rate and total damage after 100 000 years, a comparison of the results is needed. Before calculating the total damage, it is necessary to take the decay of the spent fuel into consideration. Håkansson (2000) calculated neutron dose rates for BWR (38 and 55 MWD burn-ups) and PWR (42 and 60 MWD burn-ups) assemblies as a function of time from 1 to 3×10^5 years. The results of neutron dose rates were given at two positions, outside the canister and 1 m from the canister. We select the conservative data group, that is, the one with the highest fluxes, as our reference for these calculations in order to have a conservative discussion. Figure 3-2 demonstrates the neutron dose rate ratio over time at different positions for each assembly. No discernible variation at the canister edge and outside the canister can be seen[1]. This indicates that the changes of ratio for neutron flux at different positions are the same. Furthermore, since neutron dose rate is directly proportional to damage rates, curves in Figure 3-5 can also be viewed as the damage rate ratio over time. If we integrate the data of two PWR cases from 30 to 10^5 years, we get factors of 938 and 700 for 42 and 60 MWD burn-ups respectively. Here we make a conservative choice and take 938 since it means higher damage accumulation. Thus, the total damage after 10^5 years in PWR assemblies could be calculated using the equation:

$$\text{Total dpa} = \text{dpa/year} \cdot 938 \qquad (3\text{-}8)$$

where dpa/year is the dpa rate at 30 years.

[1] The slight deviation seen for 42 MWd/kg comes from what is most likely a typo or misprint in the original data.

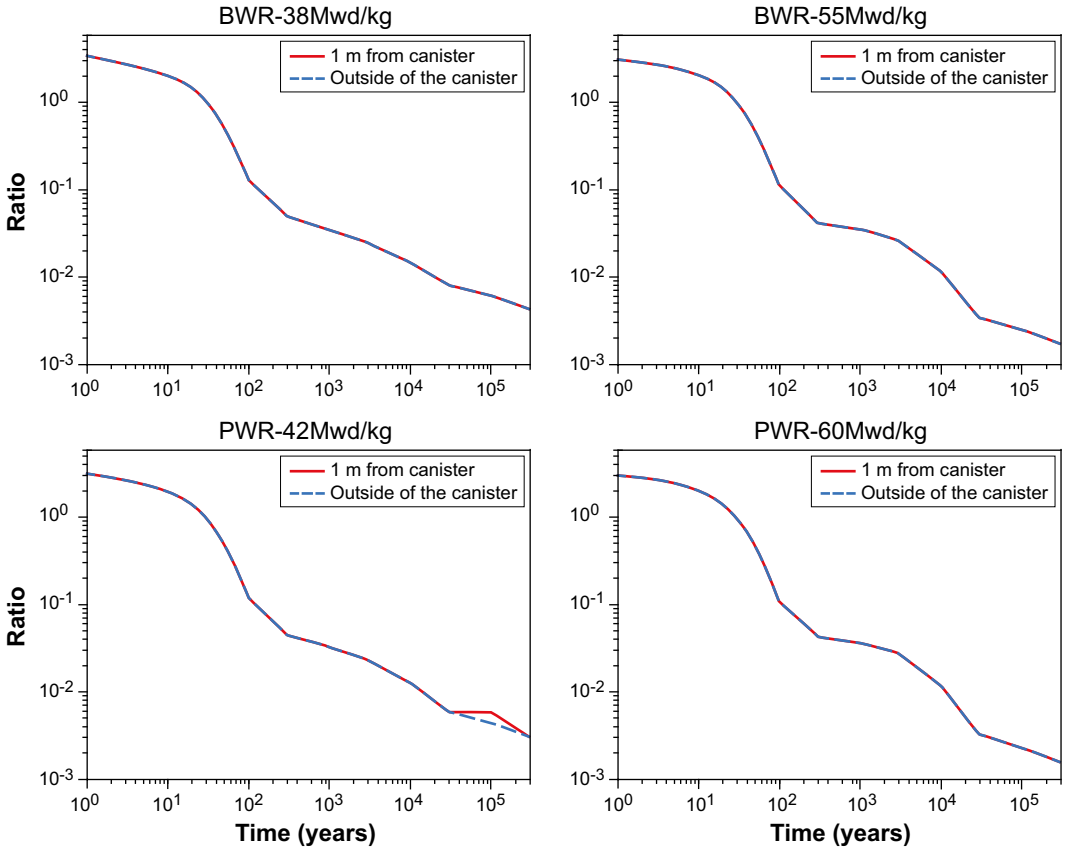

Figure 3-5. *Neutron dose rate ratio as a function of time for different assemblies. We also compared decay curves at different positions for each assembly. The ratio of neutron dose rate at 30 years is assumed to be 1 for ease of comparison.*

3.6 Comparison with reference

The total damage after 10^5 years can now be calculated. Table 3-2 shows the comparison of the dpa rate at 30 years and the total damage in copper after 10^5 years, comparing Guinan (2001) and this work. E_d for both calculations is 40 eV, for a consistent comparison. By setting up a rate theory framework, we could also calculate the amount of diffusion controlled recombination of point defects, which would lower the total amount of damage. However, as this damage rate (and total damage) is so low, the materials would always be in the extreme dilute limit with respect to the radiation damage concentration, and the mobility of defects at the temperatures in question is very low. For a consistently conservative approach, we thus assume no self-annealing. Recent progress in quantum molecular dynamics (Olsson et al. 2016) indicate a lower average threshold in Fe, but only by a factor of 0.8 and this small factor should not matter in this context. No such investigation of Cu has been done to date, however, but we do not expect any large deviation from the tabulated threshold displacement value. According to Table 3-2, it is obvious that while different codes and databases were used, the results of this work are still in good agreement with the results of Guinan (2001). In addition, since the results do not differ by any appreciable level, most conclusions regarding the effect on mechanical properties in Guinan (2001) should still be valid, as long as the irradiation enhanced precipitation of Cu in the cast iron remains at manageable levels. Recent works indicate that the chosen Cu limit of < 0.05 at. % is appropriate to avoid Cu clustering and precipitation (Chang 2015, Olsson et al. 2013). However, since there is a rough Cu/Fe interface between the canister shell and the insert, there could potentially be an irradiation induced influence from interface interactions, which may merit further study.

Table 3-2. Comparison of results at positions D, I and A for both metals between reference and this work. Both Fe and Cu were assumed to be pure. The data of Table 3-1 is used (PWR group flux, 3.5 % at 30 years).

	Guinan 2001		This work	
	Iron at D	Iron at I	Iron at D	Iron at I
Dpa/year	9.21E–10	5.15E–10	9.59E–10	5.37E–10
Dpa after 10^5 years	1.20E–6	6.70E–7	1.17E–6	6.55E–7
	Copper at I	Copper at A	Copper at I	Copper at A
Dpa/year	5.00E–10	2.92E–10	5.44E–10	3.09E–10
Dpa after 10^5 years	6.50E–7	3.79E–7	6.63E–7	3.77E–7

3.7 Production of H and He

The neutron flux interacting with materials could not only cause atomic displacements but also transmutations, in which new elements and isotopes are produced in materials. Among all new elements and isotopes, hydrogen and helium have the most pronounced influence on the mechanical properties of metal materials (Schroeder and Ullmaier 1991, Nakahara and Okinaka 1988, Fabritsievand and Pokrovsky 1997). For helium in copper, even 1–2 appm can result in considerable reduction of mechanical properties (Fabritsievand and Pokrovsky 1997). Hydrogen could also affect the mechanical properties of copper and influence corrosion processes at the canister surface if significant amounts are generated through nuclear reactions. Hence calculating the amount of H and He produced by neutron radiation after 10^5 years is important. In this section, we estimated the total amount of H and He atoms generated by transmutation using SPECTRA-PKA.

Table 3-3 demonstrates the total amount of hydrogen and helium produced in the bulk through nuclear reactions after 10^5 years at different positions for both metals. Even the maximum amount of light elements produced in both Fe and Cu are practically negligible since they are too low to reach the minimum influence level in these materials (Schroeder and Ullmaier 1991, Nakahara and Okinaka 1988, Fabritsievand and Pokrovsky 1997). Therefore, we could infer that the light elements should not affect mechanical or corrosive properties of iron or copper during 10^5 years.

Table 3-3. Total amount of H and He produced at different positions for the Fe and Cu materials.

Material	Position	Light element	dpa at 10^5 years	Total amount (appm)
Fe	D	H	2.52E–11	2.52E–5
		He	9.16E–12	9.16E–6
	I	H	1.29E–11	1.29E–5
		He	4.81E–12	4.81E–6
Cu	I	H	3.71E–11	3.71E–5
		He	9.55E–12	9.55E–6
	A	H	2.05E–11	2.05E–5
		He	5.14E–12	5.14E–6

3.8 Diffusion in copper

Irradiation enhanced self-diffusion to the outer surface of the canister could potentially lead to the formation and growth of irregular structures and surface reconstruction. Such irregularities could potentially increase the site density for corrosion or other chemical attacks. The self-diffusion coefficient in copper at 100 °C is 1.06×10^{-29} cm^2s^{-1} (Maier 1977) and the equilibrium concentration of copper vacancies at the same temperature is about 1.65×10^{-9} appm (Hehenkamp et al. 1992). The diffusion distance l could be calculated using the equation: $l = \sqrt{4Dt}$, where D is the diffusion coefficient and t is the time. The concentration of vacancies in copper at position I increased 8 orders of magnitude (from 1.65×10^{-9} to 6.63×10^{-1} appm (Guinan 2001)) due to radiation after 10^5 years and we assume that the diffusion coefficient also increased by the same factor since these concentrations are well within the dilute limit and diffusivity is proportional to the probability of finding a defect nearby (and thus proportional to the defect concentration in the dilute limit). The mean diffusion distance after 10^5 years is still only 2.32 microns, which is very small in this context and should not lead to any significant build-up of Cu atoms near the canister surface. A conservative upper limit would be a dispersed surface roughening of about a micron, if the flux of Cu atoms would be drawn strongly to the surface as a sink. This is hardly significant in the context.

4 Conclusion

New calculations of radiation damage in the copper shell of the KBS-3 canister design were here performed. For the gamma irradiation, the new calculations yield a slightly smaller damage rate in copper than previous work. The previous work can thus be considered conservative for the role of the gammas.

A different code and database for neutron damage calculations were used in this work with respect to Guinan (2001), but still the calculated damage rates are in good agreement.

In the worst-case scenario, the added gamma and neutron damage rate at 30 years is about 6×10^{-10} dpa/year, and after 10^5 years the accumulated damage should be roughly 7×10^{-7} dpa. This very low damage level should have no significant effects on the mechanical properties of the canister.

Based on our calculations, it was also found that the amount of light elements (hydrogen and helium) produced by neutron radiation after 10^5 years is practically negligible. Hence, the H and He build-up should not affect the mechanical properties of the canister materials or lead to embrittlement. For the Cu self-diffusion, since the diffusion coefficient of vacancies in Cu at and below 100 °C is very low, the calculated diffusion distance is also nearly insignificant. Thus the self-diffusion in Cu should not be a key issue either. Based on these results, we conclude that in theory radiation effects should not have any significant influence on canisters after 10^5 years.

A potential issue that has not been treated here is if there could be any radiation induced interface effects between the copper shell and iron insert.

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Arkitektkopia AB, Bromma, 2019

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