Environmental impact analysis for fusion-fission transmutation systems

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Abstract

Fusion-fission fuel cycles have been given a renewed attention in the last years as an advanced technological option for management of spent nuclear fuels (SNF) from commercial reactors, by transmutation of highly radiotoxic transuranic (TRU) elements. The use of a subcritical device for transmutation effectively increases TRU burn-up respect to the use of a critical device. In this study, performance of a small fusion facility [3], based on the Spherical Tokamak, using a fusion power <250 MW, has been evaluated. The proposed fusion device consists of a plasma fusion source surrounded by a helium-cooled fission blanket to produce a 3 000 MWth output. TRU are fed in the form of zirconium metal fuel and transmuted mainly through fission reactions driven by the fusion source. Simulations with the MOCUP coupling code showed that it is possible to transmute up to 52% of SNF TRU (burn-up: 357 000 MWd/MT-IHM) using a three-batch management strategy for the fusion device with a residence time of 2 250 days per batch. The resulting TRU material is depleted in fissile isotopes, while an increase in Cm isotopes is found. In the studied fuel cycle, TRU and new fission products are then sent to a conceptual geological repository. The repository performances are assessed in terms of radionuclide release to the biosphere per generated unit energy, comparing the fusion-fission cycle to the once-through fuel cycle. It is found that the environmental benefits of minor actinide transmutation are closely related with waste form durability, and for assumed conditions, evident only after 10^5 years.

Introduction

Fusion-fission hybrid (FFH) reactors have been studied for different purposes from the early 70s. One application of FFH that has gained a renewed attention is their use for transmutation of minor actinides for waste management [1-4]. Subcritical systems for transmutation of actinides, such as accelerator-driven systems and FFH, have been studied due to several advantages for transmutation. The main advantage of these systems is that they are not constrained by criticality and consequently the achievable burn-up for transmutation is much higher than competitive critical systems. Different studies [5-6] on the efficiency of transmutation through fusion-fission machines show how the high-level waste (HLW) radiotoxicity is greatly reduced by the additional transmutation step. However, the most important metric for waste disposal performances is the health risk of the public, such as the annual dose that could reach a target population from a repository. Therefore, the performance of the fusion-fission system needs to be expressed by such radiological safety metrics by coupling the repository source term with appropriate radionuclide transport simulations to assess the effectiveness of the fuel cycle.

In this study, a Spherical Tokamak (ST) [2,7] fusion reactor has been taken as the base for developing a FFH. The main advantage of a small fusion machine, with a ratio (Q) of produced fusion power to heating power close to 1, is that some of the material limits of a pure fusion reactor could be overcome operating close to 1 MW/m^2 neutron wall load. Since the goal of this study is to look at the fuel cycle of FFH the most important quantities of interest are the fissioned mass of actinides in a cycle and the burn-up that is reached by each fuel batch. Consequently, the results of this study can be extended to similar machines where the cycle reaches the same burn-up. In this analysis, the final radionuclide inventory produced by the system is used to determine a suitable form for waste disposal. Subsequently, the waste form is then assumed to be disposed of in a geological repository and the Transport-To-Biosphere code (TTB) [8] is used to assess the hazard at a pre-determined distance from the Engineered-Barrier-System (EBS). With this cradle-to-grave analysis, it is possible to determine the total impact on the environment of the benefits of a transmutation fuel cycle.

Reactor model and neutronics results

The geometry of the FFH reactor, considered in this study, is shown in Figure 1. The goal of this device is to transmute the transuranics (TRU) content of commercial spent nuclear fuel (CSNF). At the moment, only transmutation of minor actinides (MA) is considered. In fact, fusion devices thanks to their high-energy neutrons can effectively transmute minor actinides, while fission product transmutation would require a low-energy spectrum. The transmutation of MA is the one that can potentially have a larger impact on the reduction of radiotoxicity on the CSNF, since MA are the main contributors to long-term radiotoxicity of CSNF. The fission blanket is helium-cooled and it is divided in three zones where each batch is loaded with FA of Zr(45w/o)-TRU(55w/o) metallic fuel. The physics is essentially dominated by fast neutrons, so for calculations a homogenisation of the batch material has been done, as typical in fast reactor simulations.

For reactor simulation the MOCUP code has been used to couple the neutron transport code MCNP 1.51 and to depletion code ORIGEN 2.2. The core is initially loaded with TRU with mass fractions from CSNF. A three-batch fuel cycle is used; every 750 days a shuffling is done, charging a new batch in the inner zone of the blanket close to the plasma. The new batch is fabricated using the material of the discharged batch (after five years of cooling) and the TRU reservoir made by CSNF TRU. To simulate recycling, a recycle module was included in the MOCUP code. The reactor runs at a constant power of 3 000 MW_{th}, produced by the fission reactions. On top of this power, the fusion neutrons will deposit additional power in the blanket, slowing down.

The reference case to assess the initial TRU inventory is PWR CSNF with a burn-up of 42 000 KWh/MTU, initially enriched to 4.5% in ^{235}U, and cooled for five years. It is assumed that TRU are separated with 99% efficiency, using a PUREX process or pyroprocessing. Table 1 shows the mass fractions of TRU in the TRU mixture which constitutes the reservoir for the make-up fuel used in recycling FFH spent fuel.

Figure 1: MCNP model of FFH

Major radius = 120 cm, minor radius = 80 cm, elongation = 3.2,
fission blanket height = 250 cm, fission blanket thickness = 33 cm

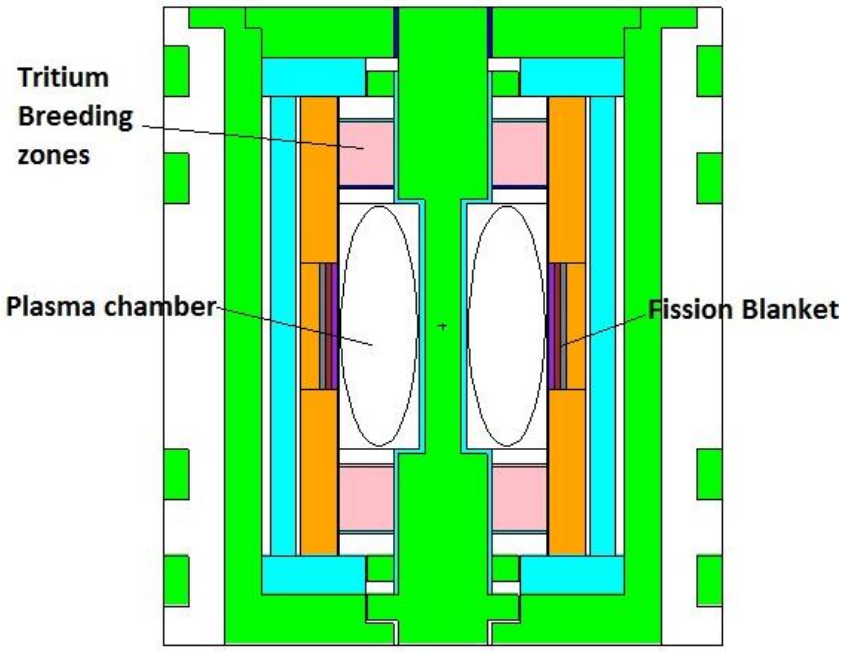


Table 1: Mass fraction of TRU from CSNF

^{237}Np	6.121E-02
^{238}Pu	1.821E-02
^{239}Pu	4.952E-01
^{240}Pu	2.391E-01
^{241}Pu	9.884E-02
^{242}Pu	4.665E-02
^{241}Am	2.937E-02
^{242m}Am	5.622E-05
^{243}Am	9.104E-03
^{242}Cm	6.537E-07
^{243}Cm	3.018E-05
^{244}Cm	2.173E-03
^{245}Cm	9.673E-05
^{246}Cm	1.226E-05

Figure 2 shows the neutronics performances of FFH where k_{eff} is calculated for a subcritical system. Figure 2 shows the system when an equilibrium state has been already reached, accounting recycling at every shuffling. The k_{eff} varies between 0.83 and 0.97 and consequently the fusion power varies between 21 MW and 237 MW. The recycling process separates 99% of TRU from the discharged batch and uses them in the new batch. Table 2 shows the TRU moles in a batch at beginning (BOC) and at the end of the equilibrium cycle (EOC); the total mass of TRU at BOC for one batch is 4.36 tonnes. Only the main TRU are shown in Table 2, though other minor actinides are contained in a batch at the equilibrium composition. The total batch burn-up is 52% (516 055 MWd/MT_{IHM}). For the purpose of this study, material issues have not been analysed and such a high burn-up might not be achievable; however a higher burn-up compares favourably to FFH cycle since it reduces the number of recycles and consequently of TRU in waste; therefore the main conclusions of this study on the hazard of FFH cycle waste remain solid. The FFH is

Figure 2: k_{eff} behaviour for FFH

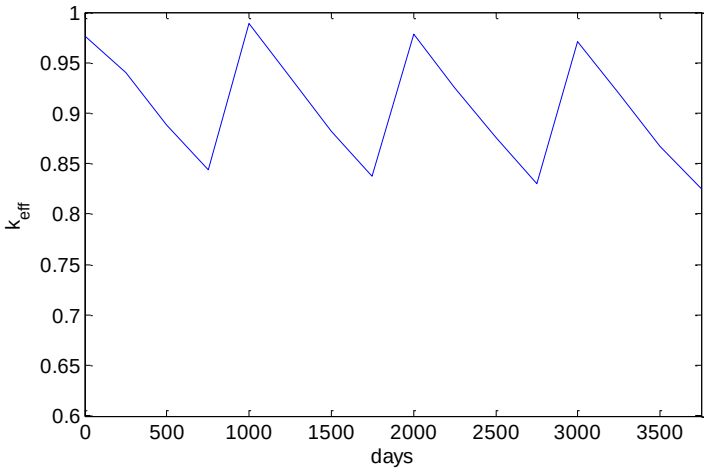


Table 2: Moles of TRU at the beginning of cycle (BOC) and end of cycle (EOC)

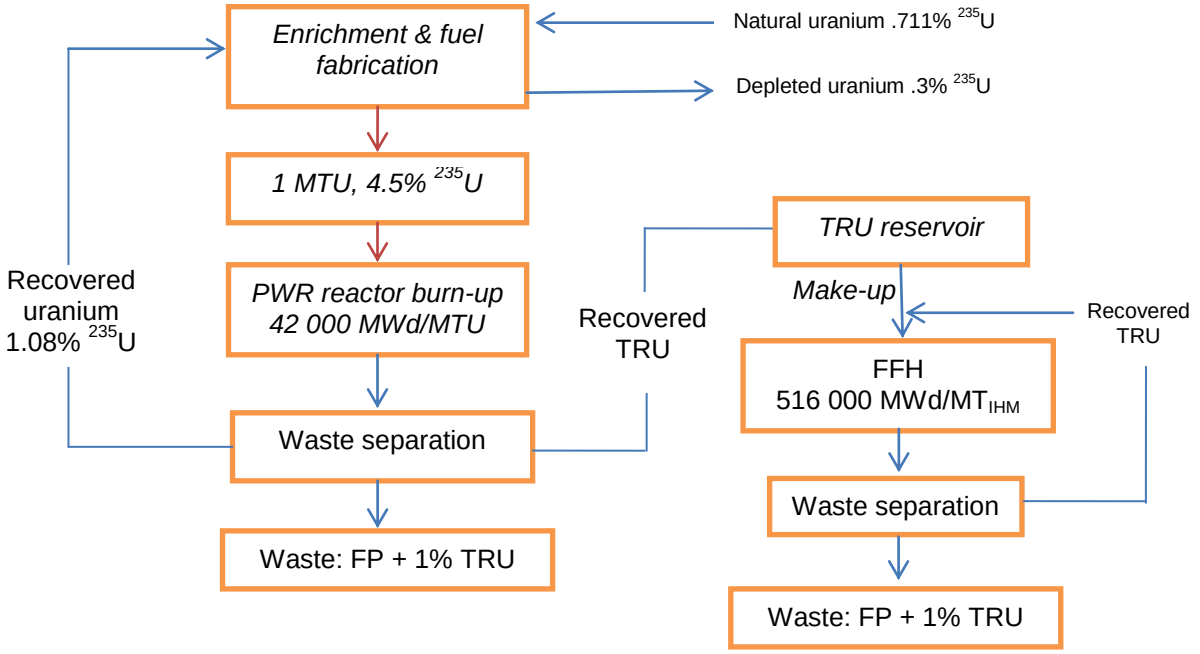
TRU	BOC	EOC
^{237}Np	8.15E+02	2.14E+02
^{238}Pu	6.66E+02	4.79E+02
^{239}Pu	6.57E+03	1.78E+03
^{240}Pu	5.99E+03	3.67E+03
^{241}Pu	1.41E+03	6.00E+02
^{242}Pu	1.47E+03	1.04E+03
^{241}Am	7.19E+02	3.05E+02
^{242m}Am	5.88E+01	5.95E+01
^{243}Am	2.61E+02	1.78E+02
^{242}Cm	1.70E-01	2.53E+01
^{243}Cm	1.64E+00	1.57E+00
^{244}Cm	1.91E+02	2.13E+02
^{245}Cm	3.45E+01	3.50E+01
^{246}Cm	1.00E+01	1.09E+01

able to transmute about 1.1 tonnes/year of TRUs which correspond to the TRU produced by approximately five PWR in a year. This result is consistent with other systems [1,3,4], so that the following calculations for the fuel cycle can be considered a good parameter for other FFH transmutation systems as well. Considering a burn-up of 52%, 1.92 passes in the FFH would be necessary to destroy completely a determined amount of TRU available as make-up fuel.

Fuel cycle

The reference PWR spent fuel contains 10.1 kg of TRU. This fuel can be burned in the fusion reactor completely; the power that FFH reactor can extract from this mass is 10 899 MWd, so that the total production of energy for the PWR-FFH fuel cycle is 52 899 MWd. Figure 3 illustrates the schematic of the PWR-FFH fuel cycle, where the CSNF is first separated in fission products (FP), uranium and TRU. The recovered uranium is still enriched at 1.08% in ^{235}U and can be recycled for fuel enrichment and fabrication. With uranium recycling and TRU burning in FFH, the fusion-fission cycle can produce 49% more energy than the once-through cycle for the same natural uranium amount used. However, it is important to notice that this estimate assumes that the uranium can be recycled infinitely at the uranium enrichment; in reality due to the increase of ^{236}U in used uranium, the neutron capture cross-sections would be too high to make a valuable use of the uranium and the recycling can be only limited to few number of times.

Figure 3: Schematic of the studied fusion-fission fuel cycle



After discharge from FFH, each batch is recycled after five years of cooling; the Zr contained in the batch is discarded and it is not recycled. During the recycling process the fission products are completely separated by the batch material while the TRU are recovered with 99% efficiency. The discarded zirconium and 1% TRU will constitute the waste of FFH which will need to be added to the waste of the first pass of recycling of PWR SNF. The burning and recycling of 10.1 Kg of TRU would produce 25.97 Kg of waste, considering that they are recycled in FFH for 1.92 times and the relative Zr is discarded at each time.

The waste produced by the recycling in FFH is compared to the waste produced by the once-through option in Figure 4. The radiotoxicity by ingestion of radionuclides is shown in m^3 of water per unit energy. It is observed how the recycling in FFH contributes to a 100 times reduction in waste radiotoxicity, contributing to shorten the time scale of the high-level radiotoxicity, since the long-term contribution of MA is greatly reduced. From this graph, it could be argued that the environmental impact of a FFH fuel cycle would be consequently 100 times smaller than the once-through option, however as shown in the following part of this study, this is not the case.

Determination of waste loading in waste form through a linear programming approach

In this section, the high-level liquid waste produced by the waste separation process is analysed to determine a suitable waste loading in the final waste form for disposal. To do so a linear programming approach is used to identify the maximum waste loading in borosilicate-glass waste form [9].

Borosilicate glass is a conventional form to solidify high-level liquid waste, as such it is assumed to be used as the waste form for the radionuclides produced from the FFH recycling. To include in this analysis all the waste generated by the fusion-fission fuel cycle, it is assumed that the waste coming from recycling FFH spent batches and from PWR spent fuel recycling can be blended in the same waste form. The waste loading in the waste form cannot be arbitrarily fixed, in fact for good vitrification of the glass waste some constraints need to be considered. The main constraints are: *i)* total waste canister mass (2 033 Kg); *ii)* volume constraints (total waste volume must be between 80 and 98% of canister volume); *iii)* molybdenum content (molybdenum mass fraction is to be less than 2% in vitrified waste); *iv)* sodium content (sodium mass fraction

Figure 4: Ingestion radiotoxicity per GWd for FFH and once-through PWR cycle

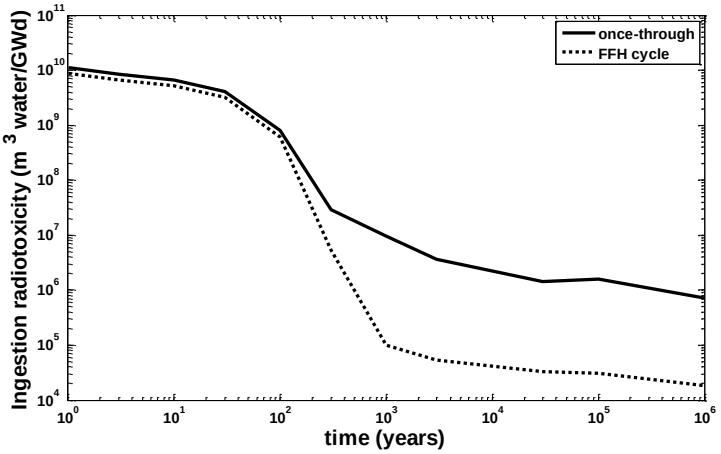
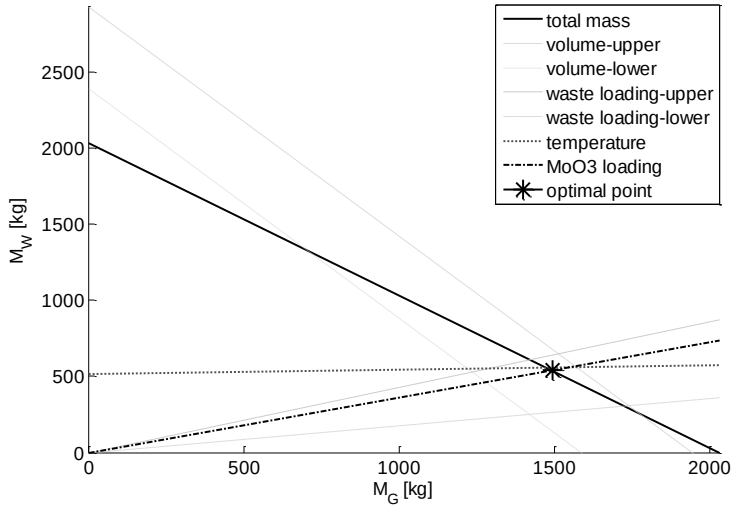


Figure 5: Linear programming approach to determine maximum waste loading

The graph shows the vitrification constraints as a function of waste mass (M_w) and the glass mass (M_G) per canister. The total mass of waste per canister is 2 033 kg. The optimal point is indicated with a star.



should be no more than 10%); *v)* waste temperature (maximum waste temperature should be less than 400°C); *vi)* waste loading (the radionuclide oxides in vitrified HLW should be between 30 and 15%). The linear programming approach [9] was applied, where all these constraints are linearised including the temperature constraint, which depends on the volumetric heat generation by the spent fuel (10.88 W/kg). For accurate complete description of the method applied and of all the constraints considered, we refer the reader to Ref. [9].

For 1 MTU of fuel for PWR, the FFH fuel cycle produces approximately 79 Kg of waste (25 of each are coming from FFH recycling). The mass of oxide HLW is calculated to be 103 Kg and it is assumed that the waste cools for 25 years before the vitrification process. Figure 5 graphically illustrates the constraints used in calculating the waste mass to be loaded in one canister. It is found that the maximum waste loading is determined by the molybdenum constraint and the total mass constraint. The waste loading (M_w) in each canister is calculated to be 540 kg and the mass of glass frit (M_G) is consequently 1 493 kg. Therefore, each canister can support the mass coming from 5.24 MTU of uranium going through the FFH fuel cycle for a total energy production of 277 334 MWd.

Radiological risk of radionuclides

The last step to assess the radiological risk of a fuel cycle is to calculate the quantity of radionuclides that are transported to the biosphere from a geological repository. To make this assessment, geological repository data about the host rock and the type of Engineered Barrier Systems (EBS) used, need to be specified. These data are taken from Table 2 of Ref. [10], where a geological repository for the Korean pyroprocessing fuel cycle is analysed.

To assess the transport or radionuclides to the biosphere the Transport-to-Biosphere (TTB) code, developed at UCBNE, is used [8]. TTB code simplifies the waste form and the EBS in two concentric spheres of same surface as the actual waste form and the actual EBS. Then, the transport of radionuclides through water in host rock fractures is calculated. For a detailed description of TTB, we refer the reader to Ref. [8]. TTB code determines, using the input solubilities, if the radionuclide of concern is released in a congruent release mode or in solubility limited mode. In particular, it is observed that most of the actinides form precipitates, since their solubility in the water is low; consequently their transport is determined primarily by the dissolution rate of the precipitates in the water than by the glass dissolution itself.

To compare the once-trough option to the FFH fuel cycle, an analysis of the inventory reaching the biosphere per canister is first done and subsequently a comparison of the two fuel cycle is carried out, normalising the radiological hazard to the unit energy produced. For the direct disposal case, it is assumed that each canister can contain 1.74 tonnes of spent fuel [10]; the FFH waste is instead a canister of 2.033 tonnes as derived in the previous description. The equivalent canister sphere has a radius r = 0.6 m, both for direct waste disposal canister and for FFH waste [9]. The EBS is assumed to be made of bentonite with an equivalent radius of 1.95 m; the host rock is granite. The dissolution time of uranium oxide for the direct disposal case is assumed to be 4 million years, given the fact that uranium oxide is a very stable form; while for the FFH waste a dissolution time of 50 000 year is assumed, which is reasonable for glass waste.

Moreover, it is assumed that the both canisters break at 1 050 years after vitrification, including the interim storage time of 50 years and the canister failure time of 1 000 years. The results on the radionuclide transport are shown below at a distance of 10 m from the EBS surface. To compare the mass-transport results in a meaningful way, the mass release rate at the 10-m location has been converted to the hazard index. The hazard index is given by the radioactivity transfer rate (Bq/year) at the determined location to the Annual Limit of Intake (ALI) in Bq/year [9]. The ALI is a measure of the hazard given for a specific radionuclide. The value for each radionuclide can be found in Ref. [9].

Figure 6 shows the hazard due to one canister for direct disposal, while Figure 7 shows the hazard from a canister of waste from FFH cycle. These figures show which radionuclides contribute to the total hazard. Knowing the energy generation from the waste of each canister, the hazard index can be normalised to the electricity generated as in Figure 8, which compares the total radiological hazard of the two fuel cycles. It is first observed that the transmutation does not reduce the hazard peak value, since this is due mainly to long-lived fission products such as ^{129}I, ^{126}Sn, ^{79}Se, ^{135}Cs. The peak happens at different times for the two waste forms, due to the different values assumed for dissolution times for uranium oxides and borosilicate glass. Figures 6 and 7 show how the release of MA such as ^{237}Np does not present any appreciable change in peak value and release time between the two different waste forms; this is due to the fact that the radionuclides have solubility limited release and consequently the precipitates that form in the proximity of the waste matrix are responsible for the release. From Figure 8 it is observed that the benefits of transmutation are apparent only after 10^5 years; however, a considerable increase of the hazard in the first part is observed which is mainly attributed to the different dissolution times for the waste forms. This implies that it is imperative to develop a durable waste form for solidification of HLLW arising from FFH recycle.

Figure 6: Hazard from one canister of PWR spent fuel

1.7 tonnes or 71 400 MWd equivalent

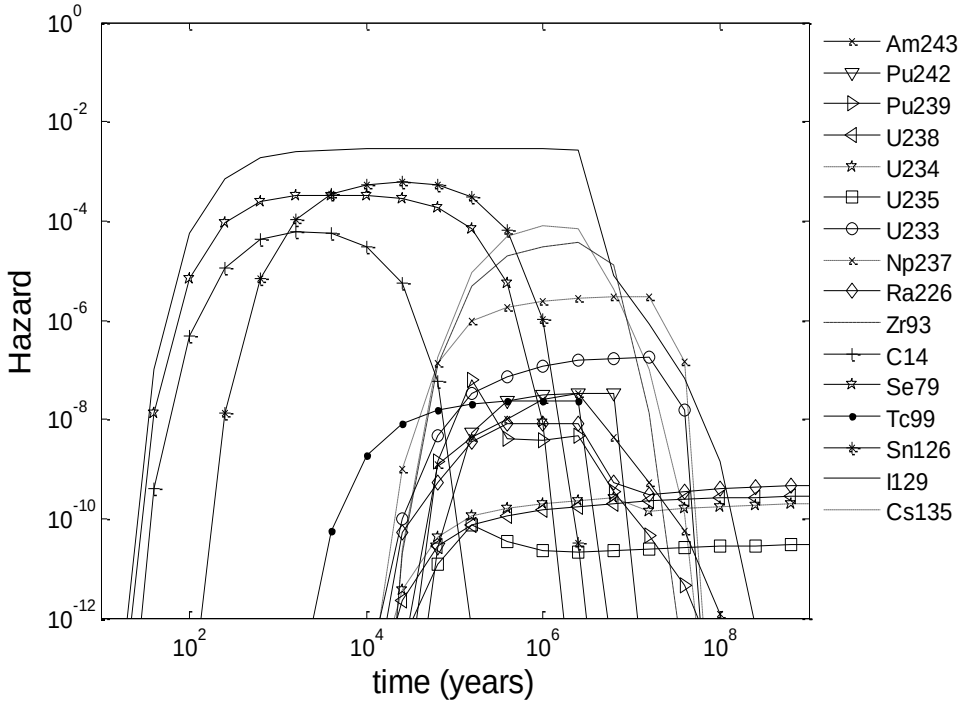


Figure 7: Hazard from one canister of FFH waste

2.03 tonnes or 277 334 MWd equivalent

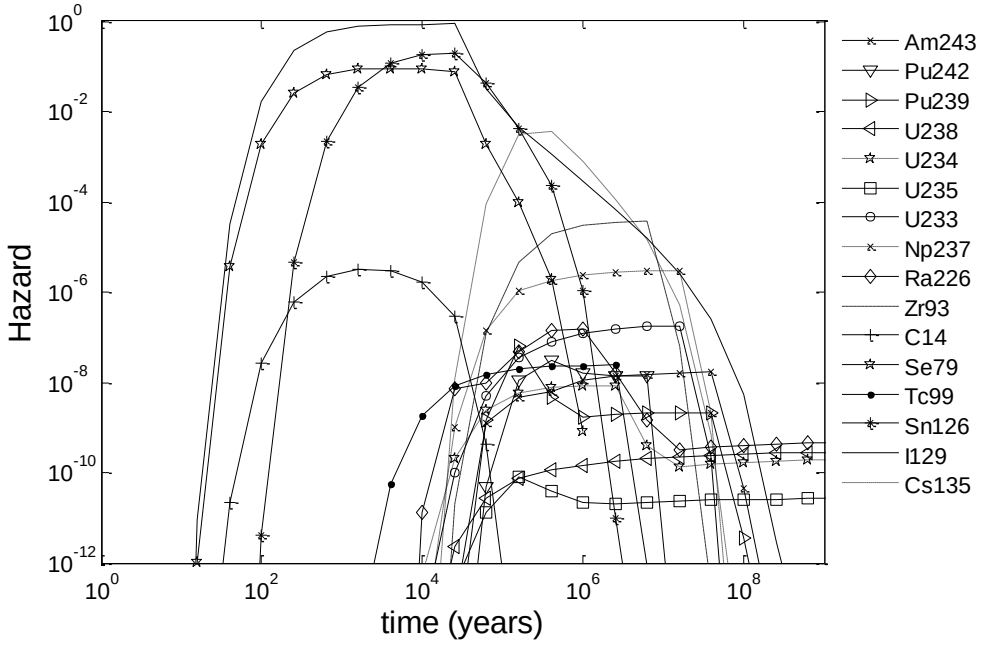


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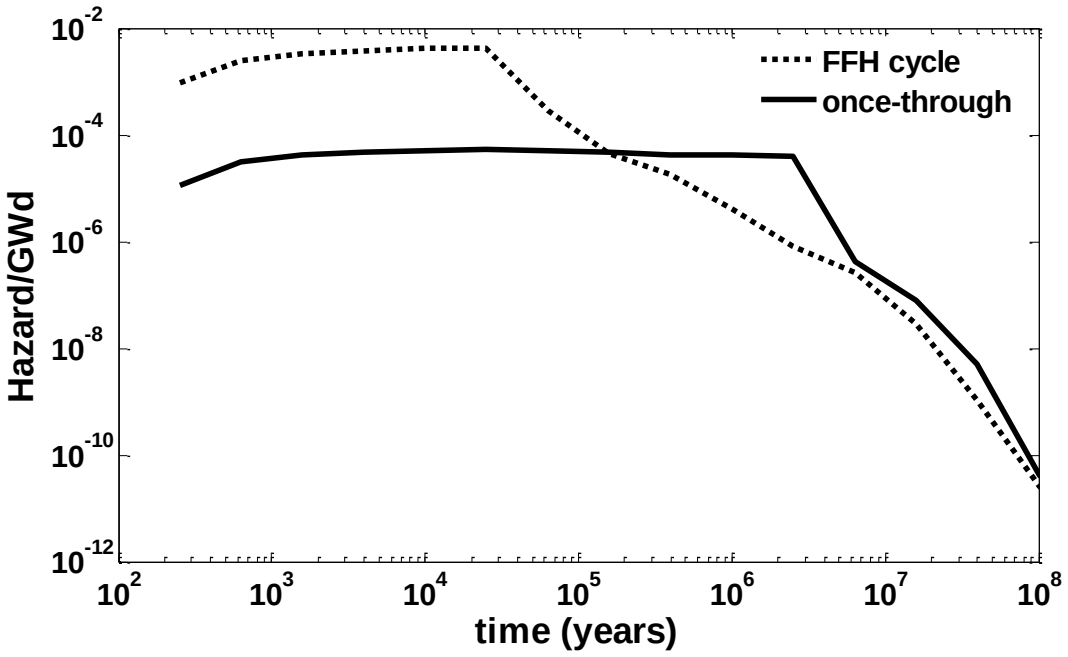
Figure 8: Comparison of hazard from FFH cycle and direct disposal



Conclusions

An analysis of a fusion-fission fuel cycle has been made, ranging from the neutronics performances of FFH to the environmental impact of the waste generated. It is shown that such a fuel cycle could produce 49% more energy that the once-through option, recovering the MA in spent fuel. An analysis of the generated waste inventory was done to identify the maximum waste loading in the waste form to use as source inventory for assessing the environmental impact of the FFH fuel cycle. It is shown that the environmental benefits from FFH fuel cycles are small in this comparison. In particular, the FFH hazard is lower than the once-through hazard only after 10^5 years, with a two-order of magnitude increase of the hazard for FFH waste in the first part of the release. To prevent this, it is imperative to develop a durable waste form for solidification of HHLW arising from FFH recycle. To effectively reduce the hazard of released radionuclides, transmutation should include also fission product transmutation which could be done for specific fission products with neutrons leaking out of the blanket of FFH. However, transmutation benefits are found in the reduction of the waste volume (one disposed canister generated 3.87 times electricity than a once-through canister) and in the increased energy production which is incremented by 49% for same natural uranium feed, which reduces generation of mill tailings and their radiological impacts. These effects should be included in the assessment in future studies.

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