The performance of Deep Burn TRISO spent fuel in a geological repository

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Abstract

The use of a Deep Burn reactor to recycle commercial spent nuclear fuel (CSNF) offers remarkable benefits including the extraction of additional electricity, added proliferation resistance and a reduction of the radiotoxicity of the spent fuel. Two central components of the Deep Burn reactor are the TRISO fuel particles, and the all-graphite core. The TRISO coatings not only allow for extremely high burn-ups, but their corrosion resistance to groundwater attack makes them ideally suited for geologic disposal. Our analysis of the performance of the TRISO particles under attack from groundwater corrosion indicate that the overall failure rate of the TRISO particles will likely be below 0.1% for between 3 000-100 000 yrs depending on the rate of dissolution of the SiC layer. The all graphite core design for the Deep Burn reactor is attractive from a repository perspective because of the slow corrosion of graphite in both air and water. Calculations of the lifetime of the graphite waste form in a flooded geological repository are in the range of 10^8*-*10^9 *years. This remarkable resistance to groundwater corrosion makes this graphite waste form a near ideal engineered barrier against the release of radionuclides from a geological repository.*

Introduction

The use of a Deep Burn high-temperature reactor to recycle light water reactor spent fuel offers the potential to extract extra electricity from commercial spent nuclear fuel (CSNF), to nearly completely destroy the ^{239}Pu present in the spent fuel, and to simultaneously transmute much of the highly toxic transuranic (TRU) content of the CSNF in to a less toxic material. An analysis of two potential Deep Burn fuel cycles show that an additional 7-16% electricity can be generated from CSNF TRU by burning them in a Deep Burn reactor. This surplus electricity generation comes without the need for any additional uranium. Furthermore, our study indicates that the Deep Burn option has the potential to reduce the total natural uranium required by 16-24%, while substantially reducing the overall radiotoxicity burden of the spent fuel by transmuting some of the more toxic elements.

Two central components of the Deep Burn Modular Helium Reactor are the TRISO fuel particles, and the all-graphite core. The TRISO particle is a fuel design that utilises three protective coatings (two layers of pyrolitic carbon with a layer of silicon carbide in the middle) to house the fissile fuel in small spheres of diameter less than 1 mm. The TRISO particles are then bonded with graphite to form cylindrical fuel compacts that are in turn loaded in to graphite fuel elements (a Deep Burn core consists of approximately 1 000 fuel elements and on the order of 10^{10} TRISO particles). The middle silicon carbide (SiC) layer is the main structural component of the TRISO particle, and acts as a miniature pressure vessel, containing the fission gasses that are created during the burning of the fuel. The TRISO coatings not only allow for extremely high burn-ups, but their robust nature with respect to groundwater attack makes them ideally suited for geologic disposal. The all graphite core design for the Deep Burn reactor is highly attractive from a repository perspective because of the extremely slow corrosion of graphite in both air and water. The direct disposal of the graphite fuel elements, therefore, represents a promising scheme for the long-term containment of radionuclides within the engineered barriers of a nuclear waste repository.

The results of an analysis of simultaneous waste package failure in an unsaturated repository are presented and CSNF is used as a basis to compare Deep Burn spent fuel (DBSF) on a GW(e)-year basis. The model allows for decay chains of arbitrary length and considers that the radionuclides can be released either congruently as the waste matrix dissolves, or in a solubility-limited mode. Because of the robust performance of graphite in groundwater with respect to the SiC layer of the TRISO particle, the protective coatings are conservatively considered to play no role in the retardation of the release of radionuclides from a failed waste package and the radionuclide inventory is assumed to be distributed homogeneously throughout the graphite waste matrix. The cases of whole element disposal and compact only disposal are considered. In both cases, the total radiotoxicity that is released into the environment seems to be substantially less than that of CSNF over geologic time periods. This robust performance is largely due to the enormous amounts of graphite that are present in the spent fuel form. This substantially reduced release rate from failed waste packages demonstrates efficacy of the Deep Burn process to reduce the overall environmental impact of CSNF.

Radiotoxicity analysis

One method for assessing the potential environmental impact of spent nuclear fuel is to calculate its radiotoxicity. Radiotoxicity is a measure of the amount of water necessary to dilute the spent fuel to the highest allowable concentration set by federal regulations. Given a fuel inventory, the computer code ORIGEN can calculate the radiotoxicity as a function of time in the repository. The radiotoxicity of a material is given as:

$$Toxicity(m^3\ water) = \sum_i \frac{\lambda_i N_i}{C_i} \tag{1}$$

where the numerator is the activity of the ith radionuclide (in Bq or Ci) and C is its maximum permissible concentration (in terms of Bq/m^3 or Ci/m^3).

There are a number of different fuel cycles and fuel compositions have been proposed for the Deep Burn process. In the following analysis, we make use of the proposed fuel loading scheme and fuel cycle found in [4] (Figure 1), along with a simple modification of the fuel cycle (Figure 2). The fuel cycle is based on a three-year residence time in the Deep Burn reactor followed by a one-time recycling of the DBSF. After removing the fission products, the TRU are combined with the Am and Cm from a LWR and the resulting fuel spends three more years in the Deep Burn reactor, after which it is transferred to an accelerator-driven subcritical reactor. The modified fuel cycle will only consider a one-pass fuel cycle based on a Np/Pu oxide fuel (with the Am and Cm from the LWR destined for long-term storage) and a single three-year burn in the Deep Burn reactor.

Figure 1: Mass flow for a one-time recycle Deep Burn process [4]

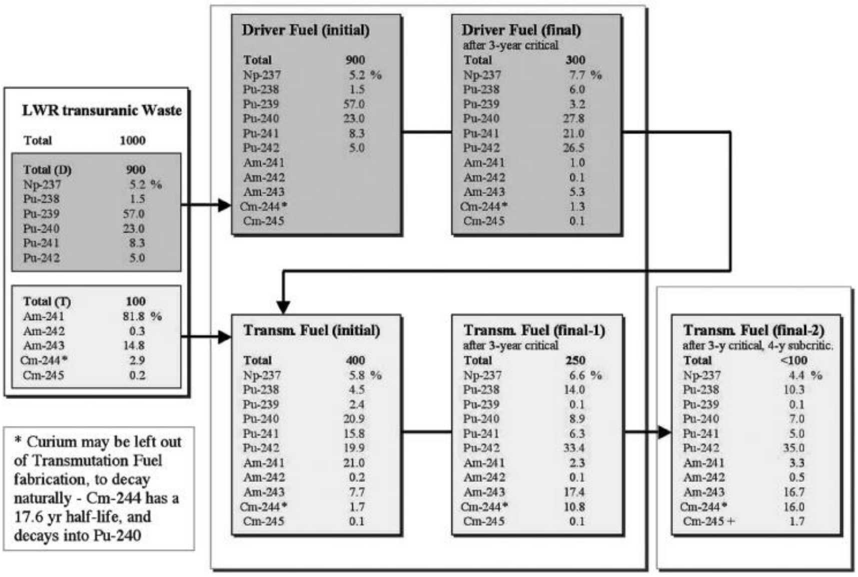
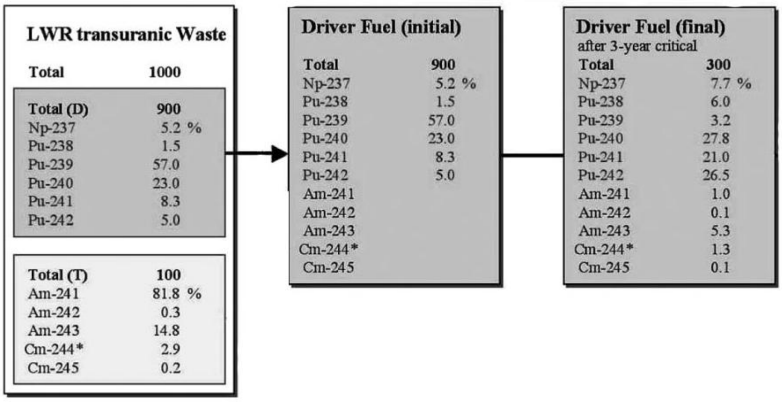


Figure 2: Once-through Deep Burn fuel cycle [4]



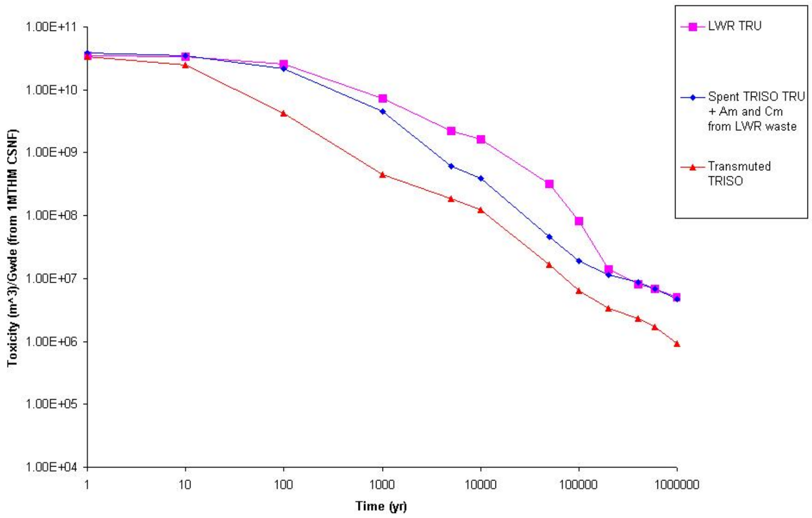
Because of their long half-lives and low maximum permissible concentrations, TRU represent the largest and longest lasting contributors to the overall radiotoxicity of a spent fuel form [10]. Therefore, as a first-order approach to quantifying the radiotoxicity of spent TRISO fuel, an analysis of the TRU content will be sufficient. By making an analysis of the content of the fuel at various stages in the fuel cycle we can draw conclusions on the effectiveness of the Deep Burn process in reducing the radiotoxicity burden in a geological repository. Below are the results of such an analysis assuming 1 MT CSNF.

The time-dependent radiotoxicity was calculated with the ORIGEN computer code. The input data is given in Table 1. As expected we observe a rapid reduction in the toxicity of the TRISO fuel in the first few years. The toxicity reduces to levels below that of the CSNF in roughly one year and remains below that level for up to the million-year point. A further reduction in the toxicity can be realised if we consider the fact that the TRISO fuel is essentially "twice utilised" fuel. That is to say, we have been able to extract more energy out of the same amount of material. Therefore, it is a fair analysis to judge the fuel forms on a GWd. Using a burn-up of 50 GWd/MTHM, a capacity factor of 0.8, and a thermal efficiency of 0.325 for the LWR and a burn-up of 600 GWd/MTHM, a capacity factor of 0.8, and a thermal efficiency of 0.5 for the Deep Burn TRISO we are able to generate the result below (Figure 3). We see that using this normalisation that the spent TRISO fuel is less toxic at the outset, and remains that way through the million-year time frame studied. Based on these results we can conclude that the one-recycle Deep Burn fuel cycle is an effective way of extracting extra energy from the CSNF while simultaneously reducing the radiotoxicity burden that the subsequent spent fuel will pose.

Table 1: LWR TRU waste and TRISO inventory from 1 MT CSNF

Nuclide	Inventory per MT LWR-CSNF							
	TRU in LWR-CSNF		Fresh TRISO		Spent TRISO		Transmuted TRISO	
	w/o	kg	w/o	kg	w/o	kg	w/o	kg
^{237}Np	4.68	0.468	5.2	0.468	7.7	0.231	4.4	0.044
^{238}Pu	1.35	0.135	1.5	0.135	6	0.18	10.3	0.103
^{239}Pu	51.3	5.13	57	5.13	3.2	0.096	0.1	0.001
^{240}Pu	20.7	2.07	23	2.07	27.8	0.834	7	0.07
^{241}Pu	7.47	0.747	8.3	0.747	21	0.63	5	0.05
^{242}Pu	4.5	0.45	5	0.45	26.5	0.795	35	0.35
^{241}Am	8.18	0.818	0	0	1	0.03	3.3	0.033
^{242m}Am	0.03	0.003	0	0	0.1	0.003	0.5	0.005
^{243}Am	1.48	0.148	0	0	5.3	0.159	16.7	0.167
^{244}Cm	0.29	0.029	0	0	1.3	0.039	16	0.16
^{245}Cm	0.02	0.002	0	0	0.1	0.003	1.7	0.017
Total	100	10	100	9	100	3	100	1

Figure 3: Radiotoxicity of CSNF, spent TRISO and transmuted TRISO normalised to total GWde produced over lifetime of the fuel



Per tonne of CSNF an additional 2.16 GWd electricity was generated for a once-through fuel cycle and up to 3.12 GWd electricity for the transmutation option, without the need for any additional raw fuel materials. This reduces our overall annual natural uranium requirement by between 0.57 MT and 1.23 MT (for once-through and one recycle, respectively) per MT of CNF. This represents a savings of between 7.7% and 16.6% on the raw materials and an additional 16.6-24% electrical power generation. Further, the ^{239}Pu inventory has been substantially reduced, thus illustrating the efficacy of the Deep Burn process as a means to mitigate the proliferation of weapons grade material.

Release model

A model has previously been developed within the Waste Management Research Group at UC Berkeley that is well adapted to use in the determination of the release rate of radionuclides from a failed waste package (or the simultaneous release from multiple failed waste packages) in a geological repository [10]. The radiotoxicity of the released radionuclides is used as a measure of the environmental impact of storing spent nuclear fuel in a geological repository. Radionuclides that are released from a failed package are considered to have an environmental impact even though some of the released material could still be contained within the engineered barriers or in the near-field host rock. In this manner we can elucidate whether DBSF is acceptable for disposal in a geologic repository, and we can use the previous study as a basis for comparison.

Although this model was developed to determine the release rates of nuclides from failed waste packages containing CSNF and defence waste, with careful interpretation it can also be applied to the case of failed waste packages containing DBSF. The main difficulty in adapting this model to DBSF stems from the disparate nature of DBSF with respect to CSNF. While CSNF was idealised as a base matrix material (UO_2 in the case of CSNF) with its radionuclide inventory homogenously spread throughout, DBSF is of a different nature entirely. It is the TRISO particle that makes the DBSF so different. Rather than having the inventory of radionuclides spread evenly throughout the matrix material, the TRISO particles contain the spent fuel in discreet locations within the graphite matrix (the fuel compact and fuel element). The protective PyC and SiC coatings of the TRISO particles prevent the release of radionuclides within the matrix material until they are compromised and the fuel kernel is exposed to the corroding environment. Additionally, the rate at which the various layers of the TRISO particle corrode (with respect to the graphite fuel compact and graphite fuel element) will likely play a key role in the rate of radionuclide release. However, if it is assumed that the graphite corrodes much slower than either the PyC or SiC layers of the TRISO particle (which is currently indicated by experimental studies), and that the TRISO particles are distributed homogeneously throughout the graphite matrix of the DBSF, then the model described previously can be applied with only minor adaptations.

Physical processes

The process of oxidation that takes place when graphite comes into contact with liquid water in the presence of air occurs with the dissolved oxygen in the water, and oxygen absorbed on the surface of the graphite, not with the water itself, while the water catalyses the reaction. This is evidenced by the fact that the oxidation rate is higher in the presence of water and the activation energy is lower [8]. Long-term degradation studies performed on graphite in a brine solution revealed the absence of hydrogen or carbon monoxide in the gas phase composition, which indicates that corrosion is not caused by the interaction of graphite with water. Further, short-term degradation studies performed on graphite in both brine and water solutions under argon, air and oxygen atmospheres show marked differences in the corrosion rates. The corrosion rates are the fastest when the graphite was under the oxygen atmosphere and slowest under the argon atmosphere. Additionally, there was a significant difference between the corrosion rates of graphite samples in brine and water under a pure oxygen atmosphere. The samples in the water/oxygen system corroded much faster than those in the brine/oxygen system. This can be explained by the fact that the solubility of oxygen in NaCl and $MgCl_2$ solutions (the two brines studied were NaCl and $MgCl_2$ based) decreases with increasing salt concentration [5]. Therefore, the corrosion rate of graphite in water will be limited not only by the availability of dissolved oxygen in the water, but also by the kinetics of the oxidation process and the solubility limit of the corrosion product(s) in the water.

When the graphite matrix of DBSF comes into contact with groundwater, the matrix begins to dissolve and radionuclides are released (assuming homogeneous distribution of radionuclides in the matrix as discussed previously). If the solubility of an individual radionuclide is sufficiently high, then that nuclide will be released congruently with the waste matrix. That is, the fractional release rate of the radionuclide will be directly proportional to the fractional dissolution rate of the waste matrix. However, if the solubility of an individual radionuclide is relatively low with respect to the waste matrix, then a low solubility precipitate of that radionuclide will form and the precipitate will dissolve at a rate governed by the mass transfer of the radionuclide into the pores of the surrounding host rock, with the concentration of the nuclide in the water adjacent to the surface of the waste form given by its solubility. Solubility limits for a number of important radionuclides are already well established [10] for certain repository conditions, however the solubility limit of graphite in water, assuming the dissolution of the graphite matrix is given by the rate of steady-state mass transfer by advection and diffusion from the surface of a cylinder, is not established. What is known about the dissolution rate of graphite is a corrosion rate that is governed by chemical reaction control. This rate of dissolution would be much greater than the rate at which mass transfer would occur by advection and diffusion.

The release of radionuclides is considered to occur by modes in this model congruent release, and solubility limited release. In the case of congruent release the fractional release rate of the nuclide is equal to the fractional dissolution rate of the waste matrix. If the solubility of an individual radionuclide is low, then a precipitate of the nuclide will form. The precipitate slowly dissolves at a rate given by the rate of mass transfer into the water in the pores in the surrounding medium, with the concentration of the nuclide in the water adjacent to the surface of the waste form given by its solubility. For the case of a graphite waste matrix the dissolution rate of the graphite is taken to be 1.3×10^{-6} g/m^2/day in the case of high solubility and 7.4×10^{-8} g/m^2/day in the case of low solubility. The full solution to the above model is available in [10].

Numerical results

Discussed in this section are the numerical results of applying the computer code for the release of radionuclides into the environment discussed in the previous sections. Tables 2 and 3 contain the necessary input data for these simulations. For all of the cases considered the package failure time was taken to be 75 000 years. The cases of high and low solubility for graphite are explored using the results of leaching experiments presented in [5]. The performance of the graphite waste matrix is compared to that of CSNF (UO_2) as a means to gauge the durability of the graphite waste form. In this analysis, it is assumed that the protective TRISO coatings are not present, and that the radionuclide inventory is spread homogeneously throughout the graphite fuel element, or fuel compact (whole element disposal and compact disposal, respectively). In the case of whole element disposal, it is considered that the entire fuel element is placed in the waste package, while in the case of compact disposal it is assumed that the fuel compacts have been removed from the fuel elements and subsequently placed into the waste package. In both cases each waste package contains the same radionuclide inventory, the difference is the smaller amount of graphite matrix in the case of compact disposal.

Figure 4 shows the fraction of remaining waste matrix in the waste package as a function of time. As can be seen in both cases the graphite fuel element and fuel compact perform surprisingly well. Even in the high solubility case more than 90% of the waste matrix (and thus the radionuclide inventory because of the homogenisation assumption discussed above) remains in the waste package for more than 10 million years. The results are even more remarkable when compared to that of CSNF. CSNF is seen to degrade completely before the 500 000-year mark.

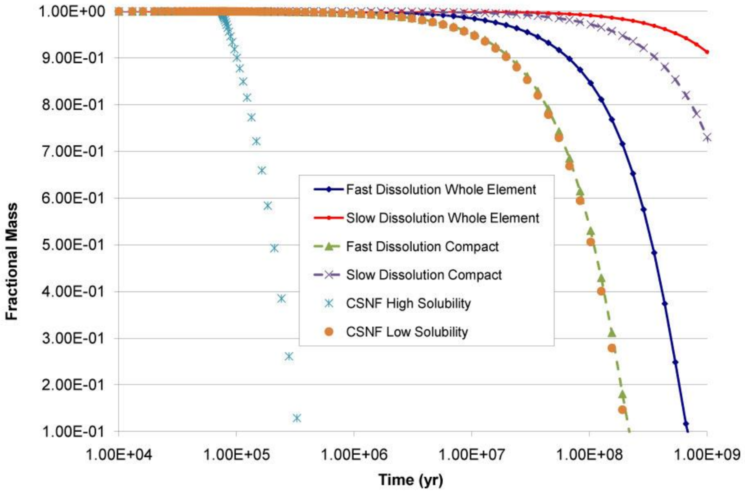
Table 2: Inventory of CSNF and DBSF (adapted from [10])

Nuclide	**Half-Life (yr)**	**MPC (Ci/m^3)**	**Inventory per package at time of emplacement in the repository (mol/package)**		**Inventory per package at time of package failure (75 000 yrs) (mol/package)**	
			CSNF	**DBSF**	**CSNF**	**DBSF**
TRU inventory						
^{245}Cm	8.50E+03	2.00E-08	9.02E-02	1.07E-01	1.99E-04	2.35E-04
^{244}Cm	1.80E+01	3.00E-08	1.29E+00	1.45E+00	0	0
^{243}Am	7.40E+03	2.00E-08	6.18E+00	6.59E+00	5.50E-03	5.86E-03
^{242m}Am	1.40E+02	2.00E-08	4.45E-02	1.29E-01	0	0
^{241}Am	4.30E+02	2.00E-08	2.71E+01	1.84E+01	1.06E-05	1.26E-05
^{242}Pu	3.80E+05	2.00E-04	2.55E+01	1.71E+01	2.23E+01	1.49E+01
^{241}Pu	1.40E+01	1.00E-06	2.32E+01	1.36E+01	3.28E-07	3.88E-07
^{240}Pu	6.50E+03	2.00E-08	3.54E+02	1.81E+00	1.19E-01	1.09E-03
^{239}Pu	2.40E+04	2.00E-08	1.86E+02	2.10E+00	2.23E+01	1.32E+00
^{238}Pu	8.80E+01	2.00E-08	7.10E+00	3.95E+00	0	0
^{237}Np	2.10E+06	2.00E-08	2.19E+01	5.085	7.06E+01	3.63E+01
DBSF waste matrix						
^{12}C	Compacts only (mol/package) neglecting TRISO coatings					8.80E+04
	Whole element disposal (mol/package) neglecting TRISO coatings					4.03E+05

Table 3: TRISO fuel, compact, element and waste package data

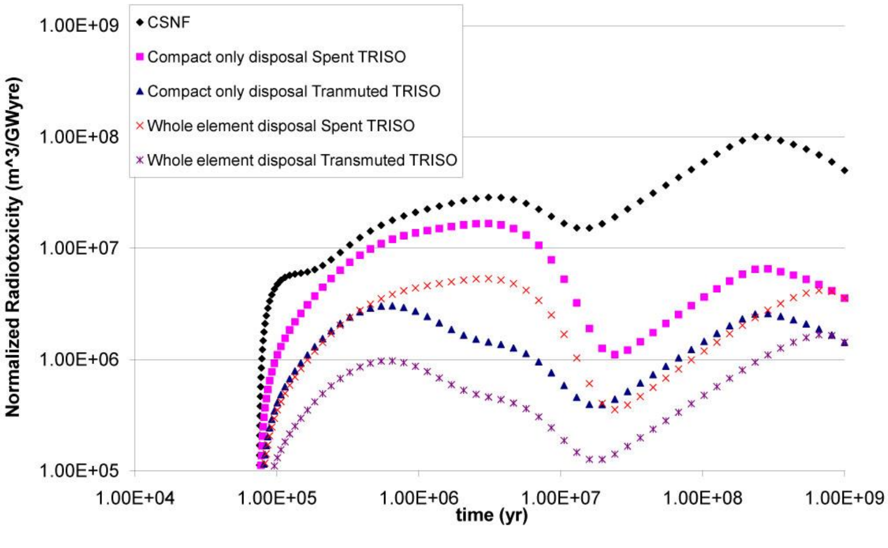
TRISO fuel	
Fuel type	TRUO2
Kernel (diameter, μm/density in g/cm^3) [12]	200/10.0
Buffer layer (thickness, μm/density in g/cm^3) [12]	120/1.05
IPyC layer (thickness, μm/density in g/cm^3) [12]	35/1.9
SiC layer (thickness, μm/density in g/cm^3) [12]	35/3.18
OPyC layer (diameter, μm/density in g/cm^3) [12]	40/1.9
Fuel compact	
Radius (cm) [12]	0.6225
Length (cm) [12]	5.114
Matrix (density in g/cm^3) [12]	1.7
Packing fraction (%) [12]	24
Number of TRISO particles per average compact [2]	10 753
Mass of fuel compact (g)	11.79
Fuel element	
Mass of graphite per element (kg) [2]	90
Dimensions (length in mm/across flats of hexagon in mm) [2]	794/360
Volume (m^3) [2]	0.0889
Number of compacts per element [2]	3126
Mass of carbon in an average fuel element including compacts (excluding TRISO coatings, kg/including TRISO coatings, kg)	115.1/123
Total mass of an average fuel element (kg)	126.9

Figure 4: Fraction of waste matrix remaining in waste package after package failure



In Figure 5 we see the results of comparing the release rates of CSNF to that of DBSF (both once-through cycle and the one-recycle option). For this calculation we considered that 63 000 MT of commercial spent fuel (the amount that is proposed for YMR) was processed and the relevant material was separated and converted into TRISO pellets destined for a Deep Burn reactor and the radiotoxicity subsequently released into the environment from failed waste packages was normalised to the total amount of energy generated over the lifetime of the fuel form. Further, we assumed the low value for the solubility of the CSNF waste matrix (UO_2) given in [10] and the high value for the constant dissolution rate of the graphite waste matrix (1.3×10^{-6} g/m^2/day). The results for the spent TRISO were generated assuming that the Am and Cm from the CSNF that was not included in the once-through fuel, was packaged into a graphite matrix in a similar manner to the DBSF. The number of waste packages is 7 886 for the CSNF, 12 073 for the once-through DBSF, and 4 829 for the once-recycled DBSF (transmuted TRISO). We can clearly see in these results that if the post-irradiated graphite does indeed perform as robustly as assumed that there is a clear benefit in terms of radiotoxicity released into the environment as well as in extra energy production.

Figure 5: Radiotoxicity in the environment from CSNF, spent TRISO and transmuted TRISO normalised to total electrical power production over lifetime of the fuel



Conclusion

From the results above, DBSF does indeed appear to be a near ideal waste form. Both the TRISO particles themselves, and the graphite fuel elements of the Deep Burn core exhibit extreme resistance to corrosion by groundwater. The TRISO coatings can provide protection to the fuel kernels from groundwater dissolution for time periods ranging from thousands of years to perhaps hundreds of thousands of years. Most remarkable, however, is the performance of the graphite. The extremely slow dissolution of the graphite offers the potential to sequester radionuclides within the waste matrix over a period of 10^8-10^9 years. Should post-irradiated graphite exhibit the same robust performance as its un-irradiated counterpart, graphite should be investigated as a potential waste form for the disposal of CNSF and other HLW.

A number of assumptions and simplifications were made that may affect the reliability of this study. First, the radionuclide inventory was taken to be homogeneous throughout the graphite waste matrix while it is in fact contained in concentrated kernels spread heterogeneously through the matrix. The release of radionuclides from failed TRISO particles could cause a deviation from the above results. Second, the experiments that provided the dissolution rates for graphite that were used in the analysis above, were not carried out on post-irradiated graphite. In order to provide an accurate estimate of the release rates of radionuclides from DBSF it is important that experiments be carried out to verify the dissolution rates that have previously been reported, or (if the material properties with respect to leaching behaviour are different for post-irradiated graphite) to provide accurate estimates of corrosion behaviour. Finally, no sorption processes with the graphite matrix were included.

References

[1] Argonne National Laboratory (ANL), *Modeling of the Repository Behavior of TRISO Fuel*, ANL-AFCI-160, Argonne National Laboratory, Argonne, IL, United States (2005).

[2] Richards, M., *GT-MHR, Assessment of GT-MHR Spent Fuel Characteristics and Repository Performance*, PC-000502, Rev. 0, *General Atomics*, April (2002).

[3] Peterson, J., M. Dunzik-Gougar, "Degradation of TRISO Fuel in a Repository Environment", *American Nuclear Society Transactions*, Vol. 90, No. 1, pp. 668-669.

[4] Rodriguez, C., *et al.* "Deep Burn: Making Nuclear Waste Transmutation Practical", *Nuclear Engineering and Design*, 222, 299-317 (2003).

[5] Fachinger, J., M. den Exter, *et al.*, "Behaviour of Spent HTR Fuel Elements in Aquatic Phases of Repository Host Rock Formations", *Nuclear Engineering and Design*, 236, 543-554 (2006).

[6] Venneri, F., "TRISO/TRIZO Corrosion (No Reprocessing)", *GNEP Final Report*, Chapter 5.

[7] Richards, M., *et al.*, "DEEP BURN Destruction of Nuclear Waste Using MHR Technology – Impact on Spent Fuel Management", *Proc. of Global 2005*, Tsukuba, Japan, 9-13 October (2005), Paper 313.

[8] Gray, W.J., "A Study of the Oxidation of Graphite in Liquid Water for Radioactive Waste Storage Applications", *Radioactive Waste Management and the Nuclear Fuel Cycle*, 3 (2), pp. 137-149, December (1982).

[9] Miller, G., R. Bennett, "Analytical Solution for Stresses in TRISO-coated Particles", *Journal of Nuclear Materials*, 206, 35-49 (1993).

[10] Ahn, J., “Environmental Impact of Yucca Mountain Repository in the Case of Canister Failure”, *Nuclear Technology*, Vol. 157, January (2007).

[11] Chambre, P.L., T.H. Pigrord, *et al.*, *Analytical Performance Models for Geologic Repositories*, LBL-14842, Lawrence Berkeley Laboratory, Berkeley, CA, United States, October (1982).

[12] “Task 2 Presentation”, *National Deep Burn Meeting*, Gaithersburg, MD, United States, April (2009).