TECHNETIUM-99 NEUTRON ABSORBERS IN THE REFLECTOR OF PB/BI COOLED REACTORS

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Abstract

The transmutation rate of ^{99}Tc as function of position in the reflector of a Pb/Bi cooled sub critical reactor has been investigated. The reactor fuel was supposed to consist of depleted uranium and transuranic waste from a light water reactor in enriched mono nitride form. The TRU fraction was varied across the core in order to obtain a flat power distribution. Monte Carlo simulations show that a maximal transmutation rate for small ^{99}Tc concentrations is obtained at 25 cm distance from the periphery of the core. The maximal transmutation rate for large concentrations is about 0.5 ^{99}Tc nuclei per fissioned TRU nucleus, if the technetium rods are placed right at the periphery of the core. This rate is more than two times higher than the technetium production rate per TRU in spent light water reactor fuel, and corresponds to usage of about half the theoretical neutron surplus in a heavy metal cooled waste transmutation system. Further, the absorption of leakage neutrons in ^{99}Tc leads to a 40% reduction of ^{210}Po accumulation due to neutron capture in ^{209}Bi.

Introduction

While ideas of transmuting long-lived radiotoxic isotopes present in spent nuclear fuel appeared over 30 years ago [1], it was not until recently that consensus appeared regarding the necessity of applying fast spectrum neutron systems for achieving sufficient transmutation efficiency of both transuranics and fission products [2-6]. Especially, it seems that applying heavy liquid metal cooling of the core would yield a neutron surplus in principle allowing for transmutation of not only ^{99}Tc, but possibly also ^{129}I and ^{135}Cs [4]. In order to evaluate the actual potential for transmutation of these fission products, one should investigate the dependence of transmutation rates on neutron spectrum at various positions in the reflector of the waste transmutation system. The influence of self shielding also needs to be well understood. In the present paper, results of Monte Carlo simulations for transmutation rates of ^{99}Tc and ^{129}I as function of radial distance from the periphery of a TRU fueled sub critical core are presented. In the following section we describe the characteristics of the waste transmutation system we have chosen as a basis for our investigations. Then we discuss the advantages of the Monte Carlo method applied for calculating the transmutation rates. Finally we display the resulting rates in various forms and discuss their implications on further studies.

Core characteristics

Fission products contribute only with a minor share of the long term radio-toxicity of spent fuel. However, due to the higher mobility of certain isotopes, transmutation of long-lived fission products would lead to a significant reduction in potential radio-toxicity leakage from a geological repository to the biosphere. Hence we have chosen to investigate waste transmutation systems with high neutron surpluses, like heavy metal cooled, accelerator driven reactors fueled by TRU waste from light water reactors (without MOX recycling). Theoretically they may yield a surplus of 1.15 neutrons per TRU fission for the purpose of transmuting fission products [4]. In practice, one has to compromise with thermohydraulics-, corrosion- and material constraints, in order to design a realistic system. Hence we limit our investigations to lead/bismuth (Pb/Bi) cooled reactors using mono nitride fuels. Methods for managing liquid metal induced corrosion on cladding and construction material have been successfully applied in Russian Pb/Bi cooled submarine reactors, while it is doubtful whether the corresponding technology for pure lead will be applicable under reactor conditions. Mono nitride fuels have been manufactured and irradiated in the US, Russia and Japan, and feature higher linear ratings than oxide fuels while being less vulnerable to radiation damage than metallic fuels. The accelerator and spallation target of our system are yet unproven technologies, but are retained in order to maximize the fission product transmutation potential. The geometry of the sub critical core used in the present investigation was set up as follows: The spallation target was assumed to have a radius of 19.5 cm. A 0.5 cm thick target wall made out of 90% Fe and 10% Cr was adopted. Five cylindrical fuel zones with a height of one meter and 10 cm thickness contained 60% (volume) Pb/Bi coolant, 10% cladding and construction material of the same composition as the target wall, and 30% "fuel". The "fuel" consisted of ^{238}U and transuranics in mono nitride form (100% enriched ^{15}N). A TRU composition similar to that of 5% heavy atom burnup PWR spent fuel after 15 years of cooling was adopted. The fraction on TRU versus U content was increased as function of radius for the threefold purpose of

- flattening the power density profile
- increasing the average neutron energy in the core for a better transmutation performance
- flattening the defect production profile in cladding and construction material.

Method of simulation

We used the continuous energy Monte Carlo code MCNP [7] for calculating multiplication eigenvalues, neutron fluxes and transmutation rates. Monte Carlo codes based on particle splitting techniques are especially useful for calculating neutron fluxes in materials with high neutron absorption cross sections, such as shielding blocks or, like in the present case, technetium rods. JEF 2.2 evaluated cross section data were processed with NJOY [8] to obtain pointwise cross section libraries suitable for MCNP at 900K (core materials) and 600K (target and reflector materials). The spallation neutron source was simulated as a volume source, with a spectrum calculated using HETC [9].

Results

First we made a reference calculation without any fission products in the reflector. The overall TRU enrichment was adjusted to obtain a neutron multiplication eigenvalue $k_\lambda = 0.96$. The fraction of TRU in the fuel zones was adjusted to obtain a flat power density profile with a ratio between maximal and minimal power density differing from unity by less than 15%. Folding the resulting neutron flux spectrum with the cross section of neutron capture in ^{99}Tc and ^{129}I, we obtained technetium and iodine transmutation rates as function of position in the reflector for concentrations small enough that self shielding effects would be negligible. Figure 1a) displays the results for technetium. As seen there is a clear maximum at 20 - 25 cm distance from the periphery of the core. This maximum is due to the combined effect of the magnitude of neutron flux declining with distance from the core, while the cross section increases as the neutron spectrum overlap with capture resonances becomes larger. In Figure 1b) the ratio of ^{129}I to ^{127}I capture rates is shown for equal concentrations of the isotopes. In the direct neighborhood of the core the probability of capture in ^{129}I is about half of that in ^{127}I. As the spectrum slows down the ratio declines to 0.3. Since isotope separation of iodine from spent fuel is hardly conceivable, we conclude that iodine should be place as close to the core as possible for efficient transmutation.

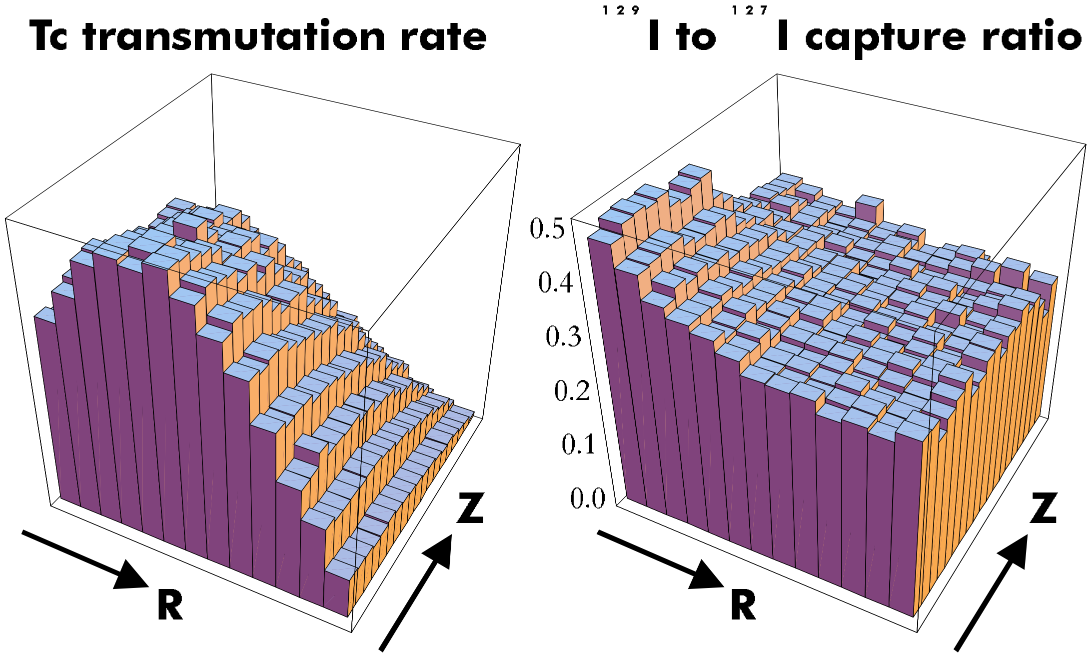

Figure 1: *The rate of ^{99}Tc neutron captures (in arbitrary units) in the reflector of the sub critical core here investigated (a). In (b) the ratio of ^{129}I to ^{127}I capture probabilities is shown. It is assumed that the concentration of the fission products is small enough for self shielding effects to be negligible. R ranges from 0 to 65 cm.*

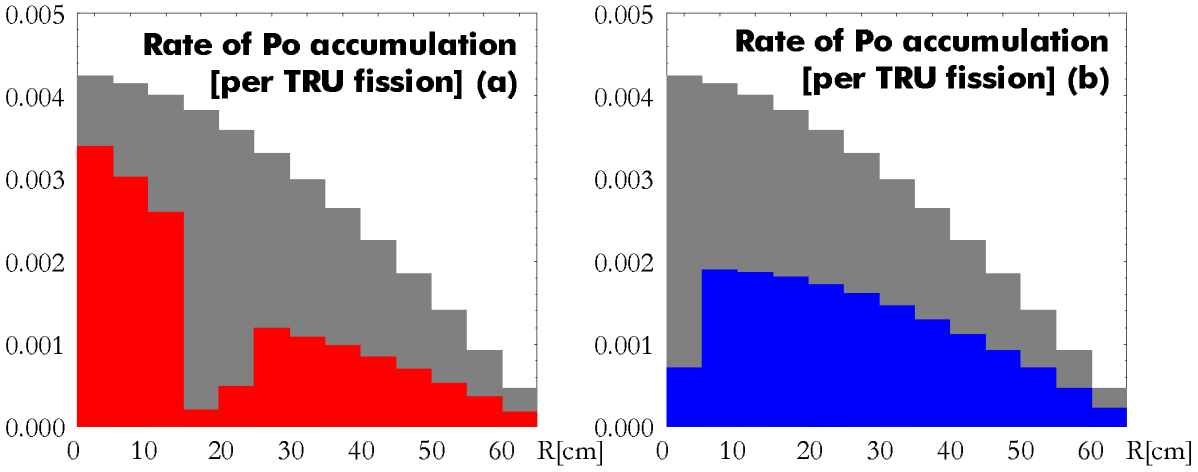

Figure 2: *The rate of polonium accumulation (per TRU fission) in the reflector of the sub critical core. In (a) 4000 kg* ^{99}*Tc was placed at a distance of 20 - 25 cm from the periphery of the core. In (b) 3100 kg was placed right at the periphery of the core. The rate of Po accumulation without any neutron absorber in the reflector is shown in the background.*

We also calculated the number of neutron captures in ^{209}Bi leading to the creation of ^{210}Po in the spallation target, the coolant and the reflector, in order to evaluate the impact of putting neutron absorbers in the reflector on global polonium accumulation.

Two calculations with ^{99}Tc added into the reflector were made. First we put 4000 kg of the substance in between 20 to 25 cm distance from the periphery of the core, corresponding to 60% of the volume in this region. The TRU content and distribution in the core was again adjusted to obtain $k_{\lambda} = 0.96$ and a flat power density profile. The number of fissions per source neutron increased slightly due to the higher concentration of TRU accompanied by the longer time needed to establish the eigenmode. It was found that 0.36 technetium nuclei were transmuted into ^{100}Ru, per fission taking place in the core. The number of polonium nuclei created in neutron captures on bismuth decreased from 0.067 to 0.047 globally, while the bismuth captures in the reflector were reduced from 0.036 to 0.016.

The bismuth capture rate as function of radius is depicted in Figure 2a) with a comparison of the rate for the technetium free reflector.

A less obvious effect was an increase in the neutron flux average energy in the periphery of the core, which can be ascribed to the fewer number of neutrons re-entering the core after slowing down in the reflector. This leads to a better transmutation efficiency of transuranic elements in the core, as the ratio between fission and capture increases with neutron energy for most of these nuclides.

Then we put 3100 kg of technetium right at the periphery of the core (0 - 5 cm distance), corresponding to 60% of the volume in this region. The number of transmuted technetium nuclei now became 0.48 per core fission, mainly due to the higher neutron flux available in the vicinity of the core. The bismuth capture rate as function of radius is depicted in Figure 2b). Polonium accumulation was not affected by moving the technetium closer to the core, but the average neutron flux energy in the core periphery was increased even further.

We also calculated the neutron energy at the point of absorption in technetium. The "capture energy" spectrum is displayed in Figure 3. Note that 98% of the captures take place at energies above 1 keV. This can to a large extent be ascribed to the effects of self shielding.

Finally, we evaluated the potential for transmuting ^{129}I by putting 230 kg of the isotope in direct connection to the core. Three tons of technetium was placed in the adjacent reflector zone. We assumed an iodine composition corresponding to a fission yield of 32% ^{127}I and 68% ^{129}I. Further, we assumed that the iodine was chemically confined as cesium iodide (natural Cs), having a comparatively high melting point (920 K). The total number of

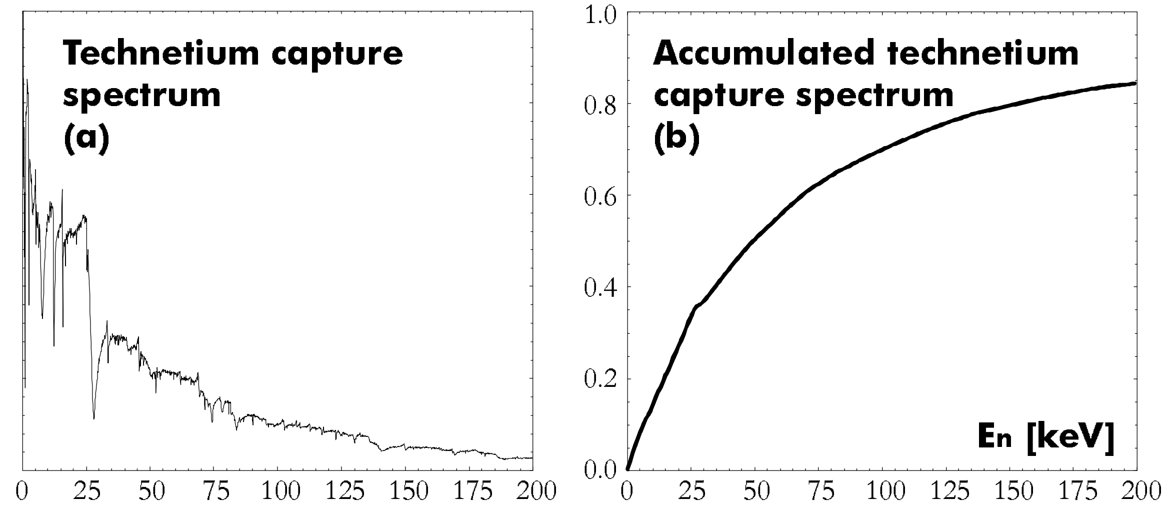

Figure 3: *Spectrum of neutron energies at the instant of technetium capture in the reflector, when having placed 4100 kg* ^{99}*Tc at R = 20 – 25 cm distance from the periphery of the core (a). In (b) the accumulated fraction of captures below a given threshold energy is given.*

transmuted fission products did not increase by much (from 0.48 to 0.50 per fission), including merely 0.024 ^{129}I nuclei. 0.060 neutrons per fissions were consumed by absorption in non toxic ^{127}I and ^{133}Cs, while the fraction of neutron captures in bismuth went down to 0.041 per fission. The transmutation efficiency of iodine appears to me much lower than that of technetium. Therefore, one should investigate if other chemical compounds of iodine may be better suited for a solid form at temperatures around 600 K. Possibly lead iodide (PbI_2) could be a candidate, if it may be produced from separated iodine at a sufficiently low cost.

Table 1 summarizes the characteristics of the four cases presented above. Note that k_s denotes the source neutron multiplication constant. For sub critical systems k_s may deviate significantly from the neutron multiplication eigenvalue k_λ, which was set to be the same ($k_\lambda = 0.96$) for all configurations.

Table 1: Summary of the characteristics of the sub critical cores investigated, assuming a total fission power of 800 MWth. All configurations had a multiplication eigenvalue equal to 0.96. The transmutation rate of ^{99}Tc (per core fission) is denoted by λ_{tr}(Tc), the ^{210}Po accumulation rate (per core fission) by λ_{tr}(Bi).

Case 1: No fission products in the reflector, $k_s = 0.953$, $\lambda_{tr}(Bi) = 0.67$

Core Zone	TRU fraction	E_n [keV]	E_{flux}[keV]	Neutron Flux [$10^{15}/(cm^2\ s)$]	PowerDensity [W/cm^3]
1	0.100	124	390	9.69	516
2	0.115	134	407	8.70	522
3	0.135	145	436	7.97	557
4	0.160	155	466	7.12	584
5	0.200	138	465	5.88	599

Case 2: Tc at R = 20 – 25 cm, k_s = 0.954, λ_{tr}(Bi) = 0.47, λ_{tr}(Tc) = 0.36

Core Zone	TRU fraction	E_n [keV]	E_{flux}[keV]	Neutron Flux [10^{15}/(cm^2 s)]	Power Density [W/cm^3]
1	0.105	128	396	10.2	544
2	0.120	138	415	9.14	550
3	0.140	150	443	8.26	575
4	0.165	163	476	7.20	582
5	0.210	163	482	5.61	562

Case 3: Tc at R = 0 – 5 cm, k_s = 0.962, λ_{tr}(Bi) = 0.42, λ_{tr}(Tc) = 0.48

Core Zone	TRU fraction	E_n [keV]	E_{flux}[keV]	Neutron Flux [10^{15}/(cm^2 s)]	Power Density [W/cm^3]
1	0.115	132	406	12.2	583
2	0.130	145	429	11.0	588
3	0.150	159	460	9.80	598
4	0.175	175	494	8.11	573
5	0.235	188	510	5.73	519

Case 4: CsI at R = 0–5 cm, k_s = 0.957, λ_{tr}(Bi) = 0.41, λ_{tr}(Tc) = 0.41, λ_{tr}(^{129}I) = 0.41

Core Zone	TRU fraction	E_n [keV]	E_{flux}[keV]	Neutron Flux [10^{15}/(cm^2 s)]	Power Density [W/cm^3]
1	0.115	134	409	11.1	603
2	0.130	146	431	9.88	603
3	0.150	158	460	8.67	603
4	0.170	173	491	7.12	558
5	0.235	187	511	4.99	514

Conclusions

In the present investigation, we showed that for small concentrations of ^{99}Tc in the reflector of a Pb/Bi cooled sub-critical core fueled with TRU waste from a LWR, the maximal transmutation rate is obtained at a distance of 25 cm from the periphery of the core. For large concentrations (60% volume) the maximal transmutation rate is about 0.5 technetium nuclei per core fission, if the neutron absorber is placed right at the periphery of the core. As a beneficial side effect, the overall polonium accumulation is decreased by 40%. Further, the average neutron flux energy in the periphery of the core is increased by 10%, which yields a better transmutation efficiency of the TRU:s in that region.

As the relative fraction of technetium in spent light water reactor fuel is about 0.2 Tc nuclei per TRU nucleus (~6% per fission), all configurations here investigated are able to transmute LWR technetium at higher rates than that of their concomitant TRU destruction rates. The transmutation efficiency of iodine here found is slightly too small for keeping up with its production rate. However, as the present system utilizes less than half of the theoretical neutron surplus in a heavy metal cooled sub critical reactor, there will be room for improvements in the design, especially so with respect to choice of iodine compound.

Acknowledgments

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References

[1] M. Steinberg, G. Watsak and B. Manowitz, Neutron burning of Long Lived Fission Products for Waste Disposal, BNL-8558, Brookhaven National Laboratory (1964).

[2] T. Sasa et al, in Proceedings of the international conference on future nuclear systems, Global 97, 425 (1997).

[3] M. Salvatores et al, in Proceedings of the international conference on future nuclear systems, Global 97, 428 (1997).

[4] M. Salvatores et al, in Proceedings of the international conference on future nuclear systems, Global 97, 561 (1997).

[5] F. Venneri et al, Disposition of Nuclear Waste using Sub Critical Accelerator Driven Systems, LA-UR 98-985, Los Alamos National Laboratory (1998).

[6] C. Rubbia et al, Fast neutron incineration in the Energy Amplifier as alternative to geologic storage, CERN/LHC/97-01, CERN (1997).

[7] MCNP - A general Monte Carlo N-Particle transport code, version 4B, LA-12625-M. Editor J.F. Briesmeister, Los Alamos National Laboratory (1997).

[8] R.E. MacFarlane and D.W. Muir: The NJOY nuclear data processing system, versions 97-99, Los Alamos National Laboratory (1998).

[9] IAEA study on the neutronic potential of a modular fast spectrum for radiotoxic waste transmutation, editor I. Slessarev, CEA/Cadarache (1998).