PHYSOR 2004 -The Physics of Fuel Cycles and Advanced Nuclear Systems: Global Developments
Chicago, Illinois, April 25-29, 2004, on CD-ROM, American Nuclear Society, Lagrange Park, IL. (2004)

PBMR Deep-Burn: a pebble-bed high temperature gas-cooled reactor burning its own "waste"

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A broader acceptance of nuclear power as a solution for the increasing energy demand by our society will be directly linked to the solution of the radioactive waste issue. Lately a lot of effort has been dedicated on studying ways to minimize the amount of waste and to decrease the time span it should be stored safely in deep geological formations.

The work discussed here utilizes the Deep Burn concept applied to the 268 MWth PBMR design to incinerate and transmute long-lived transuranic elements in a single burn-up cycle. The reactor is fuelled partly with transmutation pebbles loaded with the transuranic elements originating from discharged UO_2 fuelled pebbles used in the same reactor. The results show that about 75% of the initial amount of transuranic waste is being incinerated, while the time span in which the radiotoxicity of the waste surpasses that of uranium ore is reduced to a third. The fissile component of the plutonium disappears almost completely (incineration of more than 99%). A total power of 58 MWth is produced by the transmutation pebbles, with a consequent 25% decrease in the amount of fresh UO_2 fuel needed.

KEYWORDS: HTR, Deep-Burn, Transmutation, Waste, Pebble Bed, Actinides

1. Introduction

Unless society accepts a drastic reduction of its standards of living, energy consumption will keep on increasing and therefore the need for large-scale energy sources, which at the same time can meet the strong requirements regarding its impact to our environment. At this pace, the renewable sources of energy like solar, wind, and biomass will not be able to fulfill the requirements of the present and future societies. In the last years there is a growing awareness that nuclear power can fulfill this role as a large scale energy source with a limited impact to the environment, certainly considering a minimum emission of greenhouse gases associated to it and the relative reduced amount of waste produced. Some of the elements in this waste are radioactive and represent a hazard to life and therefore should be isolated safely in an interim storage until their radioactivity decreases to acceptable levels. To limit the burden to future generations the amount of hazardous waste should be minimized as well as the time it should be isolated from the biosphere.

The main source of public concern towards the nuclear waste and its storage in deep geological formations is the long time that it should be stored safely. This is the main issue that motivates the present research on partitioning and transmutation. Lately, the issue of sustainability has also gained further importance in relation with the Generation IV program, where one of the requirements is the development of future reactors that can utilize the

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available resources more efficiently and minimize the production of waste.

Presently, light water reactors (LWR) can re-utilize part of the discharged fuel: uranium and plutonium. Especially plutonium is of major concern because its contribution to the toxicity of the waste dominates over that of the other elements present in the radioactive waste. However, in this kind of reactors the consumption of plutonium within one cycle is limited by the maximum burnup that can be achieved with its type of fuel. With the present technology, the only option to increase the incineration of plutonium is a multi-recycling fuel cycle, with the consequent production of waste during each reprocessing and irradiation cycle.

Gas-cooled high temperature reactors (HTR) offer unique opportunities to improve the transmutation of actinides. These reactors utilize fuel elements based on the TRISO ceramic-coated-particles technology, manufactured in the shape of graphite pebbles or blocks. These fuel elements can be irradiated up to very high burnup levels, which presently cannot be reached for any other fuel type. In the past, plutonium coated particles have been successfully tested to burnups as high as 750 MWd/kg in irradiation experiments performed in the Peach Bottom [1] and Dragon reactors. Moreover, this type of fuel works as waste confinement in the final repository because of its excellent characteristics for confining fission products and actinides.

Within General Atomics in San Diego, the idea emerged to give the HTR a double function: generation of useful heat and the burning of several actinide elements. In their study [2], this is referred to as Deep Burn and it is applied for a block type GT-MHR.

The study presented in this paper focuses on this double application in a pebble bed HTR. Whereas in the General Atomics study the actinides to be burnt are taken from LWR waste, the philosophy of this approach is to use the reprocessed actinides from the same reactor: the actinides produced in the pebble bed reactor are redirected into the same reactor for a second life. The main goal is to analyze how the amount of actinide in the waste could be reduced in this way, and to evaluate its effect on the evolution of waste radiotoxicity. A preliminary cost analysis of this scenario [3] shows that there is also a cost advantage of operating the reactor in Deep Burn mode in industrialized countries, where the waste storage fees charged per volume are relatively high.

2. Model and General Assumptions

The work presented in this paper concentrates on the 268 MWth (110 MWe) PBMR design [4], which is a high temperature helium cooled reactor of the pebble bed type and utilizes a direct closed Brayton cycle for the energy conversion system. The general characteristics of this design are summarized in Table 1. The reactor core is divided in two zones: a central zone filled with graphite pebble and an annular zone filled with UO_2 fuel pebbles. This design is of the recirculation type, where both the fuel and the graphite pebbles are continuously fed at the top of the bed and discharged at the bottom. When a fuel pebble is discharged, its burnup is measured. If its maximum burnup has not been reached it is fed back at the pebble-bed top, otherwise it is discarded and a fresh pebble is loaded instead.

In the fuelling mode considered here the fuel zone is still filled with the original fuel elements (9g heavy metal (HM) mass and 8.1% enrichment) and to be referred to as driver fuel (DF). However, the central zone (20% of the total reactor core volume) is filled with elements with transmutation fuel (TF). The TF pebbles are loaded with the actinides coming from the discharged driver fuel after a reprocessing step to separate the fission products and the uranium isotopes. In this scenario, the fission products are sent to a repository and the uranium either to a repository or to an enrichment plant for re-utilization.

Table 1 Main parameters of the PBMR in Deep Burn mode

Thermal power (MWth)	268
Electric power (MWe)	110
Core height (m)	7.54
Outer radius (m)	1.75
Central column radius (m)	0.79
Volume of driver zone (m^3)	57.8
Volume of transmutation zone (m^3)	14.7
Number of driver fuel elements	312000
Number of transmutation fuel elements	79000
Packing fraction	0.61
Radial reflector thickness (m)	1.5
Top/Bottom reflector thickness (m)	2.5/3.5

Table 2 shows the main parameters associated to the driver and transmutation pebbles. The TF pebbles are loaded with 1.5g HM with an enrichment of about 45% (with ^{239}Pu and ^{241}Pu as the major contribution). The TRISO coated particles containing transmutation fuel are manufactured with kernels with a smaller diameter (240 μm) than the UO_2 ones (500 μm). The main reason for this choice is twofold: (1) with smaller kernels the resonance absorption contribution is enhanced and consequently the transmutation of non-fissile isotopes is improved; (2) a larger burnup limit can be achieved when the volume of the porous buffer zone is increased (improved volatile fission product confinement).

Table 2 Main parameters for the pebbles containing driver and transmutation fuel

	Driver Fuel	**Transmutation Fuel**
Fuel type	UO_2	$(Np,Pu,Am,Cm)O_x$
HM loading (g/pebble)	9.0	1.5
Enrichment (%)	8.1	45
Diameter of fuel kernel (μm)	500	240
Coated particles per pebble	15000	22500
Max. burnup (MWd/kg)	80	746
Residence time in core (years)	3	8.7

The flow of driver and transmutation fuel through the core is matched in such a way that the reactor is operated at a constant power, with the transuranic actinides (TRU) from the used driver fuel being reprocessed and redirected into its fuel cycle. The discharged pebbles containing TF once reaching their maximum burnup are sent to a repository without further reprocessing. At the beginning of the reactor operation, no inventory of TRU is available to load the central reactor zone. The reactor will be operated in the original PBMR mode, with UO_2 fuel and graphite pebbles, for about 10 years. During this period the fuel pebbles irradiated up to 80 GWd/tHM will be stored in order to cool down and reprocessed afterwards. Considering an inventory of TRU of about 0.13g per reprocessed driver pebble, about 11 driver pebbles are needed to manufacture one transmutation pebble. After this initial saving period, the transmutation pebbles are then fed continuously in the central zone of the reactor. In this way a new equilibrium core is achieved with 20% of transmutation pebbles and 80% of driver pebbles. A period of about 8.7 years is required to save enough quantities of TRU to fill the central transmutation zone, which is also the residence time of a transmutation pebble in the core when equilibrium condition is achieved.

3. Reactor Physical Model

3.1 WIMS model

The model used in the study has been implemented in the modular code WIMS [4], using WSNAP as the main module to determine the flux and power distribution, and the core reactivity. This module solves the multigroup diffusion equation in several geometry's using the finite difference method. In this case the (r,z)-geometry option has been used. Figure 1 sketches the cylindrical model implemented.

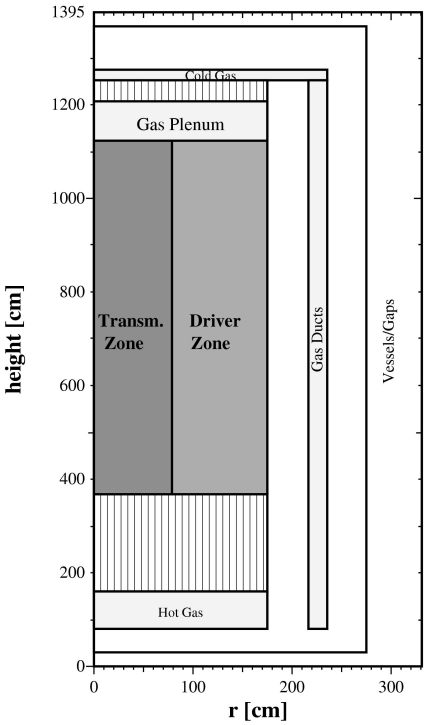

Fig. 1 Calculational model of the 268 MWth PBMR design

A total of 62×260 (radial×axial) meshes has been applied, with a typical mesh spacing of about 6.0 cm.

The nuclear data in two energy groups used in WSNAP have been generated using a 172-groups library based on JEFF2.2. Different materials (with different nuclear data) are associated to each of the reflector and fuel zones defined in the model. The generation of these nuclear data has been performed in two steps:

a- 1-D models to generate data for the zone filled with driver fuel. The data for driver fuel zone is generated under the assumption that the central column is filled with graphite pebbles.

b- 1-D models to generate data for the central column filled with transmutation fuel. These models utilize the nuclear data for the driver fuel at average burnup generated in the first step, and therefore these data do not change during the depletion calculation.

In each of these steps two separate 1-D models have been implemented in order to take into account the difference in neutron spectrum in the two-group nuclear data structure: (1) a cylindrical pseudo-reactor model to generate data for the pebble bed and the radial reflector materials; (2) a slab pseudo-reactor model to generate data for the axial reflector zones and for the pebble bed zones right adjacent to the top and bottom reflectors. The 1-D calculations are performed in a 16-neutron-energy-groups structure, using the collision probability method.

For the unit cell calculation in 172 energy groups, the double heterogeneity of the pebbles is taken into account by considering an equivalent cylindrical cell containing the TRISO coated particles. The collision probabilities for this cylindrical cell are calculated and then applied by another module to determine the flux distribution. Before applying the nuclear data to the cylindrical cell in the above mentioned 1-D models, the fine-groups constants are collapsed into the 16-groups structure, followed by an averaging of the cross sections over the detailed spatial dependence of the flux in the unit cell.

To allow for the determination of the nuclear constants and the isotopic composition in the fuel grains as a function of the burnup, the burnup module WBRNUP is integrated in the 1-D

pseudo-reactor calculation scheme. For each burnup step, a new unit cell calculation is carried out using the new isotopic composition.

3.2 FISPACT model

In the scenario considered here the discharged transmutation pebbles are supposed to be sent to a repository after reaching its maximum burnup level. At this point we will refer to the discharged fuel as radioactive waste. In order to assess the radiotoxicity of this waste, the inventory code FISPACT [5] has been used in this study. Besides calculating the inventory of nuclides in a certain material after irradiation with a given neutron field, this inventory code can also determine the potential biological hazard of the irradiated material. As source for the effective dose coefficients it uses data from the ICRP-72. The results for the radiotoxicity shown in the following sections will relate exclusively to the radiotoxicity of the actinides as result of intake by ingestion.

4. Results

4.1 Composition of Discharged Waste

The results obtained from the simulation of the depletion of the transmutation fuel show that the heavy metal content after the irradiation period of 8.7 years is reduced by 75%: from an initial 1.5 g/pebble to about 0.38 g/pebble. The plutonium amount (the main contributor to the radiotoxicity of the discharged waste) is destroyed by as much as 87%: from an initial 1.33 g/pebble to about 0.17 g/pebble. Analysis shows that Pu and Np are being burnt, at the expense of substantial production of Am and Cm. Table 3 shows the isotopic composition of actinides (in grams and weight percentage) per transmutation pebble at the beginning and at the end of its lifetime.

Table 3 Isotopic composition of the transmutation fuel at the start (80 MWd/kg) and at the end of irradiation (746 MWd/kg) per year

	Initial (80 MWd/kg)		Final (746 MWd/kg)	
	wt%	m [g]	wt%	m [g]
U233	0.00	0.0	0.00	0.0
U234	0.00	0.0	0.35	12.2
U235	0.00	0.0	0.09	3.1
U236	0.00	0.0	0.08	2.9
U237	0.00	0.0	0.00	0.0
Np237	4.00	544.2	0.64	22.0
U238	0.00	0.0	0.00	0.0
Pu238	1.63	222.1	2.85	98.0
Np239	0.00	0.0	0.00	0.0
Pu239	35.50	4831.8	0.42	14.4
Pu240	29.57	4024.6	1.52	52.2
Pu241	10.21	1390.2	0.66	22.8
Am241	5.75	783.3	0.01	0.5
Pu242	11.85	1613.4	40.04	1378.8
Am242m	0.01	0.8	0.00	0.0
Cm242	0.00	0.0	0.02	0.8
Am243	1.29	174.9	18.88	650.2
Cm243	0.00	0.5	0.00	0.1
Cm244	0.18	24.3	33.28	1145.9
Cm245	0.01	0.9	1.16	39.8
Total	**100.00**	**13611**	**100.00**	**3444**

The fraction of the fissile Pu isotopes (^{239}Pu and ^{241}Pu) is reduced by more than 99%, whereas the amount of the higher isotope ^{242}Pu is hardly changed. The isotopes with the largest concentrations in the discharged waste are ^{242}Pu and ^{244}Cm. Especially ^{244}Cm is of considerable importance since it produces a substantial amount of heat by α decay (2.5 W/g) and it has a high probability for spontaneous fission, producing neutrons. This will have consequences for the transport and storage of the waste within a period of about 50 years. Within this period the ^{244}Cm decays to its daughter ^{240}Pu.

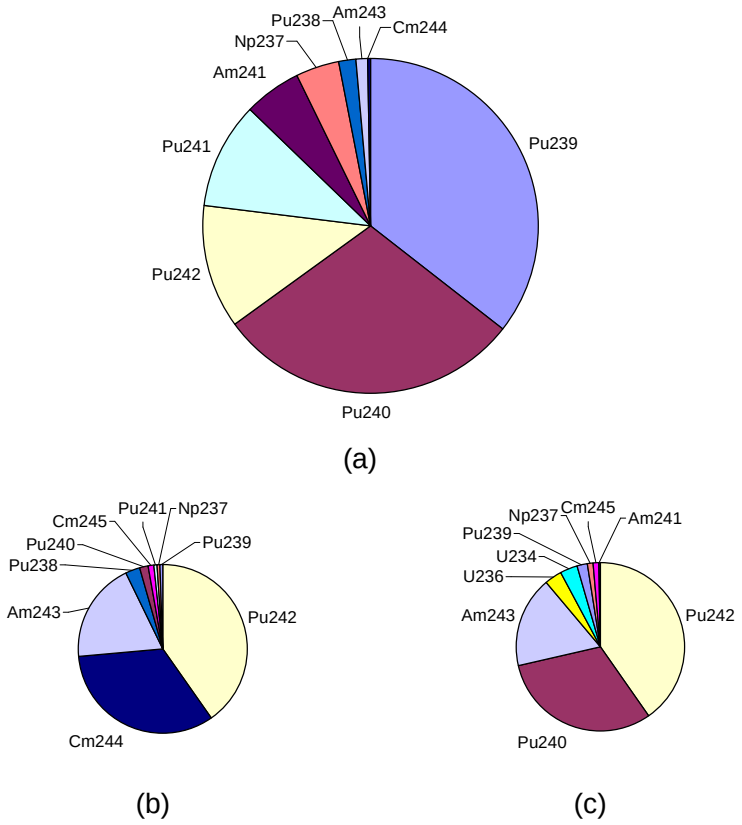

Fig. 2 Evolution of the isotopic composition of the transmutation fuel: (a) before irradiation (80 MWd/kg), (b) after irradiation (746 MWd/kg), and (c) 1000 years after disposal in the repository site. The area of each chart is directly proportional to the fuel amount.

In a different scenario the reprocessing of this waste could be considered in order to extract curium (with ^{244}Cm as the main component). After a cool-down period of about 50 years this consists almost entirely of ^{240}Pu and could be utilized as a non-parasitic burnable poison in another reactor type. ^{240}Pu has a half-life of 6550 years, and therefore plays an important role in the radiotoxicity of the discharged waste (see Fig. 4) as will be shown in the following section. This alternative scenario will not be further discussed in this paper.

Although the amount of Am and Cm isotopes increase as result of their irradiation, the results show that 2.3% of the power produced in the transmutation zone (1.3 MW) originates from fission of these isotopes. When the Am and Cm isotopes are not included in the transmutation fuel, the results show an increase of 60% in the amount of Am and Cm produced per year. Moreover, separation of only Pu+Np from the irradiated driver fuel for the

manufacture of transmutation fuel could be a proliferation concern. Therefore, transmutation of Pu and the minor actinides altogether gives the best prospect for the reduction of waste.

The evolution of the amount and vector of the transmutation fuel before and after irradiation are illustrated in Figure 2. The area covered by the charts is directly proportional to the amount of fuel. Also displayed is the status after a period of 1000 years in storage, as calculated with FISPACT. The destruction of the Pu isotopes and the breeding of the higher actinides, like ^{244}Cm and ^{243}Am are clearly visible. After 1000 years, the full amount of ^{244}Cm has decayed to ^{240}Pu, and an amount of ^{234}U and ^{236}U has originated from α decay of the relatively short-lived Pu isotopes: ^{238}Pu and ^{240}Pu. Moreover, the amount of ^{239}Pu has increased by a factor of five because of the large amount of ^{243}Am present in the discharged waste.

4.2. Radiotoxicity of Discharged Waste

The ingestion radiotoxicity inventory of the waste as function of time is shown in Figure 3. Only the actinide contribution is considered in this figure. Two curves are included, representing two scenarios: (a) Deep-Burn scenario where the transmutation fuel is irradiated in the reactor up to 746 MWd/kg and (b) original PBMR scenario where the driver fuel irradiated up to 80 MWd/kg is sent directly to the final repository without further reprocessing. Also shown is the reference radiotoxicity level of the required amount of natural uranium, in equilibrium with its daughter products.

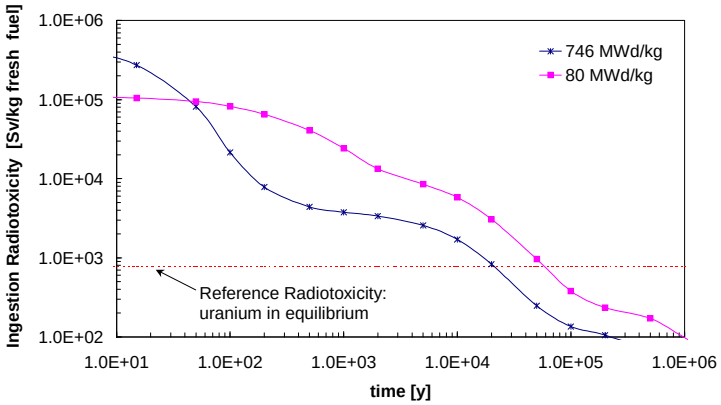

Fig. 3 Actinide radiotoxicity inventory in irradiated fuel as function of time. Results are included for the Deep Burn scenario and the original PBMR scenario

It can be noticed from this figure that the radiotoxicity of the discharged waste in Deep Burn mode is higher than in a standard PBMR scenario during the first 50 years of storage. The surplus is attributed to the large contribution of ^{244}Cm bred during the irradiation period. However, this increase in radiotoxicity would become less important if the contribution of the short-lived fission products had also been taken into account.

After this period the radiotoxicity decreases substantially, mainly because of the effective destruction and transmutation of the Pu isotopes when irradiated as transmutation fuel in Deep Burn mode. As consequence the time period necessary for the radiotoxicity of the waste to reach the level of natural uranium is reduced to a third: from about 60000 years to 20000 years. Although the plutonium inventory has been substantially incinerated during the 8.7 years of irradiation, its contribution to the total radiotoxicity still prevails over the other actinides, with ^{240}Pu as the main contributor (see Fig. 4). As discussed above, its main source is the large amount of ^{244}Cm.

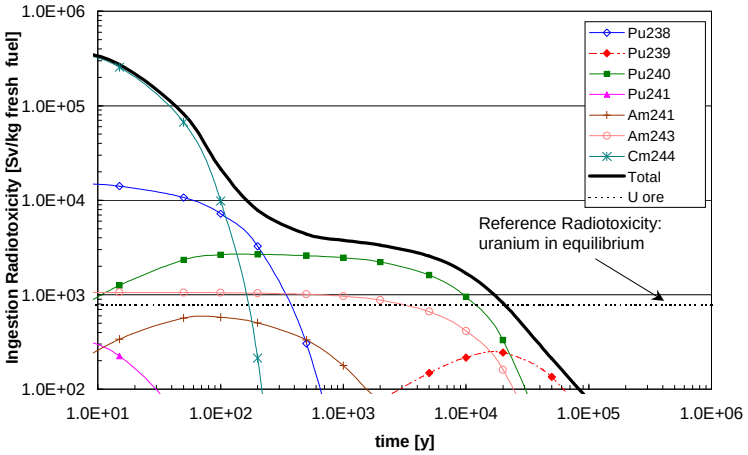

Fig. 4 Contribution of the most important isotopes to the total radiotoxicity of the waste from the PBMR in Deep Burn mode

4.3 Reactivity and Power Distribution

At this stage of the study the recirculation of the pebbles has not been modeled. As an approximation for the equilibrium situation the two fuel zones have been filled with pebbles with the average burnup level, namely 40 MWd/kg and 370 MWd/kg for the driver and transmutation pebbles, respectively. Table 4 below includes the main results obtained with the WSNAP calculations.

Table 4 Main parameters for the PBMR DeepBurn at steady-state conditions

	Driver Zone	Transmutation Zone
k-effective	1.0223	
Power (MW)	210	58
Neutron flux (10^{18} m^{-2} s^{-1})	1.38	1.54

4.4 Temperature Coefficient of Reactivity

The temperature coefficient has been calculated for the equilibrium condition at the operation temperature of 700°C. The partial contributions of the two fuel zones and of the reflector have also been determined, by varying the temperature separately and uniformly within each of these zones. These results are shown in the Table 5, where it can be seen that the temperature coefficient for the reactor is negative.

Table 5 Temperature coefficient of reactivity for the full reactor, and the partial contributions of each fuel zone and the reflector. $\alpha_T = \Delta(1/k)/\Delta T$

	α_T (pcm/K)
Full reactor	-1.1
Driver zone	-0.3
Transmutation zone	-1.4
Reflector	+0.4

5. Conclusions

The transmutation characteristics of the 268 MWth PBMR design operated in Deep Burn mode have been analyzed. In this concept the fuel cycle has been closed, with a single recycling of the discharged fuel. The core is divided in two fuel zones with a volume proportion of 1:4 (inner/outer zone). The outer zone is loaded with fresh UO_2 fueled pebbles, which are irradiated up to 80 MWd/kg. When discharged these pebbles are stored for cooling and afterwards reprocessed to extract the TRU elements. These so-called transmutation pebbles are loaded in the inner core zone, and are manufactured using the TRU elements discharged from the outer zone. At equilibrium conditions a transmutation pebble remains in the core for a period of about 8.7 years, and the results show that they can reach a burnup of 746 MWd/kg. The discharged transmutation pebbles are sent to a repository without further reprocessing. The initial amount of TRU is reduced by 75%, with an incineration of 87% of the plutonium content. The isotopic composition of the waste changes considerably: the fissile component of plutonium is reduced for more than 99%, whereas higher actinides (especially ^{244}Cm and ^{243}Am) are created. With the incineration of the TRU, the time span needed for the radiotoxicity of the waste to reach the radiotoxicity of uranium ore is reduced to a third: from 60000 to 20000 years. The contribution of ^{240}Pu, originated mainly from α decay of ^{244}Cm, dominates the total radiotoxicity from about 1000 years onwards.

A total of 58 MWth is being produced by fission of the transmutation fuel, and consequently the amount of fresh driver fuel is reduced by as much as 25%.

Acknowledgements

The authors wish to thank R.A. Forrester for making EASY-2001 available for this study.

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