THERMAL LOADING OF A REPOSITORY FOR VITRIFIED HIGH-LEVEL RADIOACTIVE WASTES FROM REPROCESSED LWR SPENT FUEL INCLUDING PLUTONIUM-RECYCLING, AND FROM FBR SPENT FUEL

W. Hauser, H.W. Wiese
Kernforschungszentrum Karlsruhe GmbH
Institut fuer Nukleare Entsorgungstechnik
Institut fuer Neutronenphysik und Reaktortechnik
Postfach 3640, D-7500 Karlsruhe 1
Federal Republic of Germany

W. Wenzel
D.T.I. Dr. Trippe Ingenieurgesellschaft mbH
Amalienstr. 63/65, D-7500 Karlsruhe 1
Federal Republic of Germany

ABSTRACT

According to the German concept, vitrified high-level radioactive wastes from reprocessed spent light-water reactor fuel elements are intended to be disposed of in a salt dome. The decay heat of this waste causes a significant temperature increase in the waste form itself and in the surrounding rock formation. This thermal loading of the host rock is an important factor of long-term safety assessments for nuclear waste repositories.

In this paper the calculated heat release of high-level radioactive wastes from reprocessed uranium dioxide and uranium/plutonium mixed oxide fuel elements from light-water reactors is discussed. In addition a comparison is given to the heat release of high-level wastes from reprocessed spent fast breeder reactor fuel elements. For the calculations the KORIGEN-code was used with input data for reference-type reactors. The calculation of the time dependent radionuclide composition of the high-level wastes was based on element separation factors for the reprocessing which were experimentally determined.

GENERAL BACKGROUND

In the Federal Republic of Germany, vitrified high-level radioactive wastes (HLWC) from reprocessed light-water reactor (LWR) fuel elements are intended to be disposed of in a salt dome. In this waste the decaying radionuclides cause a release of heat which increases the temperature in the waste form and in the surrounding salt rock formation. This results in geochemical and geomechanical effects which increase with the thermal loading of the host rock. Important effects are for example the migration of brine inclusions along a temperature gradient (1) and the induced stress and creep in the rock salt (2). Therefore, long-term safety analysis for the nuclear waste repository requires detailed information about the heat generation of the vitrified HLWC and the thermal loading of the repository.

Heat generation of the HLWC can be characterized by the thermal power and the cumulated heat depending on time. Both values depend on the irradiation history of the fuel and the type of reactor. In the planned Wackersdorf reprocessing plant uranium dioxide (UO2) and uranium/plutonium mixed oxide (MOX) fuel elements from LWRs are intended to be reprocessed. Alternatively or in addition to a plutonium-recycling in thermal reactors, it is possible to insert the reprocessed plutonium in fast breeder reactors (FBR) with a subsequent reprocessing of core and axial blanket material in a future FBR reprocessing plant.

In this paper the calculated heat release of vitrified HLWC from reprocessed LWR and FBR spent fuel elements is discussed. In detail, a comparison of thermal loadings of the repository caused by HLWC from LWR-UO2, LWR-MOX and FBR fuel elements is performed. This has been done to obtain basic informations on the thermal loading of repositories for different concepts of plutonium recycling.

For the calculations the KORIGEN-code (3) was used with consistent determination of reactor specific neutron cross-sections. Input data were the composition of the fuel elements, reactor core design and operation characteristics of reference-type reactors. Furthermore, reference element separation factors of HLWC concerning the separation of fission products and actinides during reprocessing have been considered.

REFERENCE-TYPE REACTOR FUELS

In the Federal Republic of Germany most of the operating nuclear power stations are light-water reactors. In order to use simple calculation methods the Biblis pressurized water reactor with 1300 MW electrical power was taken as a reference-type LWR. In the investigations an average fuel burnup of 40 GWd/THM was considered. The corresponding residence time of the fuel elements in the reactor core was taken as 1000 days in three cycles of full-power reactor operation.

For the charge of the reactor core five possibilities were investigated. In a first step (a) 100 % UO2 fuel, initially enriched to 3.6 % U-235 was assumed. The corresponding composition of the fuel is listed in Table I.

A reprocessing of spent fuel elements gives the possibility to recycle the reprocessed plutonium. In light-water reactors this is done by replacing UO2 fuel elements by uranium/plutonium mixed oxide (MOX) fuel elements. In model (b) a reactor core consisting of 90 % UO2 fuel (initial

TABLE I

Composition of LWR Fuels (g/THM);
Reference - Type Reactor: Biblis

	LWR core charged with UO_2 fuel	LWR core charged with UO_2 and MOX fuel		
	UO_2 fraction 100	UO_2 fraction 90% ; 70%	MOX fraction, 1. Pu-recycling step, 10% ; 30%	MOX fraction, 5. Pu-recycling step, 10% ; 30%
U 234	2.736E2	2.736E2	5.320E1	5.320E1
U 235	3.600E4	3.600E4	7.000E3	7.000E3
U 238	9.637E5	9.637E5	9.465E5	9.288E5
Pu238	---	---	6.967E2	1.365E3
Pu239	---	---	2.763E4	2.795E4
Pu240	---	---	1.129E4	1.833E4
Pu241	---	---	4.366E3	7.051E3
Pu242	---	---	2.462E3	9.423E3
Pu_{fiss}	---	---	3.200E4	3.500E4
Pu_{tot}	---	---	4.644E4	6.410E4

enrichment 3.6 % U-235) and 10 % MOX fuel containing natural uranium and 3.2 % fissile plutonium of the first Pu-recycling generation was considered (fuel composition see Table I)(4).

The common reprocessing of spent UO2 and MOX fuel which is foreseen in the German Wackersdorf reprocessing plant is limited by a MOX fraction of 30 % (4). Model (c) was based on this balanced MOX fraction with a reactor charge of 70 % UO2 and 30 % MOX fuel with the compositions given in Table I.

Furthermore, the case of multiple plutonium recycling is included. Earlier work (4) gave an equilibrium composition of MOX fuel after the fifth recycling step. This composition (Table I) was taken for model (d) with 10 % and model (e) with 30 % MOX content of the reactor core.

As a reference-type fast breeder reactor the French Superphenix 1 with about 1300 MW electrical power was chosen. The reactor core was assumed to be charged with first generation LWR plutonium (Table II). In the calculations the residence time of the fuel elements (core and axial blanket material) was 640 days in two cycles of full-power reactor operation. The fuel was assumed to be discharged after a burnup of 62.7 GWd/THM for the inner and outer core zones, and 3.15 GWd/THM for the axial blanket. This results in an averaged burnup of 40.5 GWd/THM (averaged over the core and the axial blanket).

REPROCESSING DATA AND HLWC CHARACTERIZATION

According to the concept for the Wackersdorf plant a reprocessing of all types of spent LWR fuels seven years after reactor discharge was considered. The calculations were based on the HLWC element separation factors which are listed in Table III. These element separation factors were derived from experimentally determined data for dissolver residues (5). It was assumed that the vitrified HLWC contains 0.5 % U and 0.5 % Pu-losses related to the original U, Pu-fuel inventory.

In case of reprocessing of spent FBR core and axial blanket material in a future FBR reprocessing plant a shorter cooling time of one year was considered. Table III shows the corresponding HLWC element separation factors with the same U, Pu-losses as in case of HLWC from LWR fuel reprocessing. The element separation factors of the fission products were taken from earlier investigations (6).

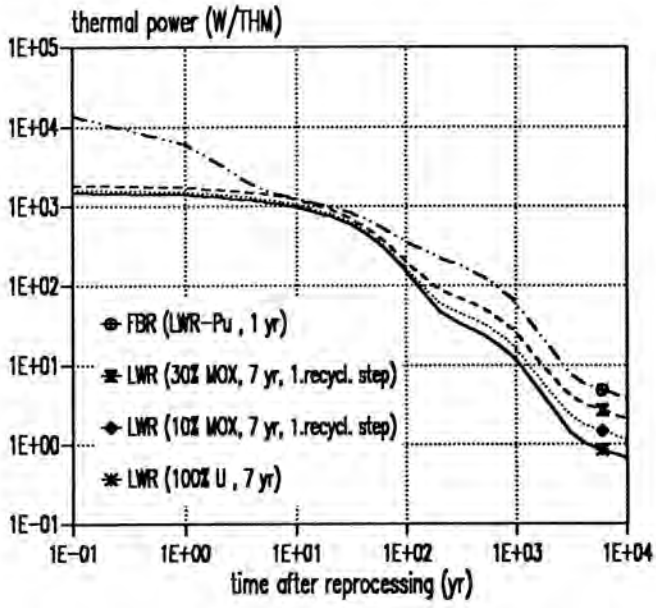
RESULTS

Vitrified HLWC is planned to be disposed of in a geological salt formation about 1000 m below ground level. After disposal of the wastes the maximum stored thermal energy in the repository is reached in a period of about 10000 years (7). After this time the thermal power which is released from the HLWC is lower than the thermal power which is dissipated by convection at the surface of the earth.

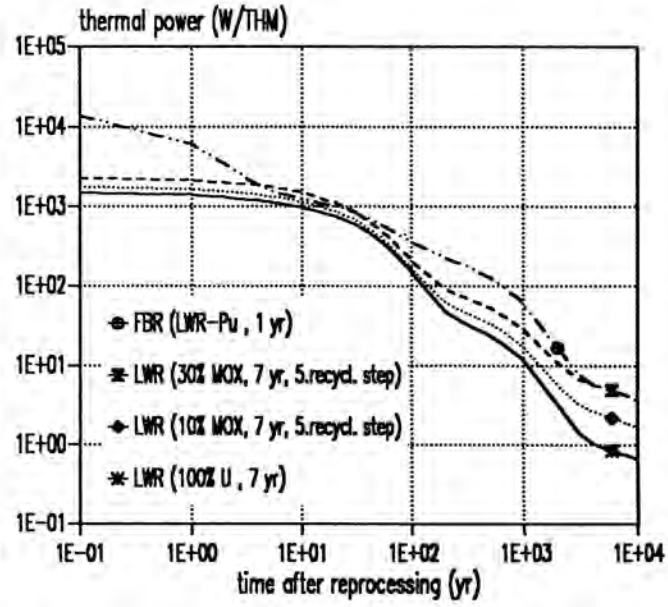
TABLE II

Composition of FBR Fuels (g/THM);
Reference-Type Reactor: Superphenix 1

	FBR core charged with first-generation LWR plutonium fuel		
	inner core zone fraction 33.2%	outer core zone fraction 29.5%	axial blanket fraction 37.3%
U 235	3.82E3	3.63E3	4.49E3
U 238	8.38E5	7.95E5	9.95E5
Pu238	2.67E3	3.40E3	---
Pu239	8.92E4	1.14E5	---
Pu240	3.73E4	4.76E4	---
Pu241	1.73E4	2.21E4	---
Pu242	8.94E3	1.14E4	---
Am241	2.54E3	3.24E3	---
Pu_{fiss}	1.07E5	1.36E5	---
$Pu_{tot.}$	1.55E5	1.99E5	---



(a) 1. Pu-recycling step of LWR MOX fuel



(b) 5. Pu-recycling step of LWR MOX fuel

Fig. 1. Thermal Power of Vitrified HLWC from Reprocessed Spent LWR and FBR Fuels.

TABLE III

HLWCElement Separation Factors (%) Related to the Original Fuel Inventory

	element	ordinal number	HLWC from LWR fuel reprocessing	HLWC from FBR fuel reprocessing
fission products	Ge	32	99.90	99.90
	As	33	99.90	99.90
	Se	34	59.90	99.90
	Br	35	99.99	99.99
	Kr	36	0.00	0.00
	Rb	37	99.99	99.99
	Sr	38	99.99	99.99
	Y	39	99.79	99.99
	Zr	40	64.65	68.65
	Nb	41	4.99	99.99
	Mo	42	49.95	51.95
	Tc	43	79.95	53.95
	Ru	44	39.80	42.80
	Rh	45	49.82	34.82
	Pd	46	79.94	63.94
	Ag	47	4.99	99.99
	Cd	48	79.99	99.99
	In	49	99.99	99.99
	Sn	50	19.90	99.90
	Sb	51	4.93	67.93
	Te	52	59.90	99.90
	J	53	0.01	0.01
	Xe	54	0.00	0.00
	Cs	55	99.99	99.99
	Ba-Er	56-68	99.99	99.99
actinides	Tl-Po	81-91	100.00	100.00
	U	92	0.50	0.50
	Np	93	99.50	99.50
	Pu	94	0.50	0.50
	Am	95	99.29	99.99
	Cm	96	99.99	99.99
	Bk	97	100.00	100.00
	Cf	98	100.00	100.00

Therefore, a relevant time period of 10000 years was chosen for the investigations on the thermal loading of a repository.

Thermal Power

Fig. 1 shows the calculated thermal power of vitrified HLWC from reprocessed spent LWR and FBR fuels as a function of time after reprocessing. These data are based on one metric ton heavy material (THM) charge to the reactor. According to the decay of the radionuclides the thermal power of the different types of HLWC decreases by a factor of 2000 to 3000 in 10000 years. In the period below about 100 years the thermal power is dominated by the fission products. The main contribution above some hundred years is given by the actinides. The calculations demonstrate that the lowest thermal power is produced by HLWC from reprocessed spent LWR uranium fuel. The largest thermal power is released by HLWC from reprocessed spent FBR fuel. Until a period of 10 years this difference results from the shorter cooling time of 1 year before reprocessing of the spent FBR fuel. During this time it contains more short-lived fission products. Above about 100 years a higher content of actinides (e.g. Am-241) causes the highest amount of thermal power.

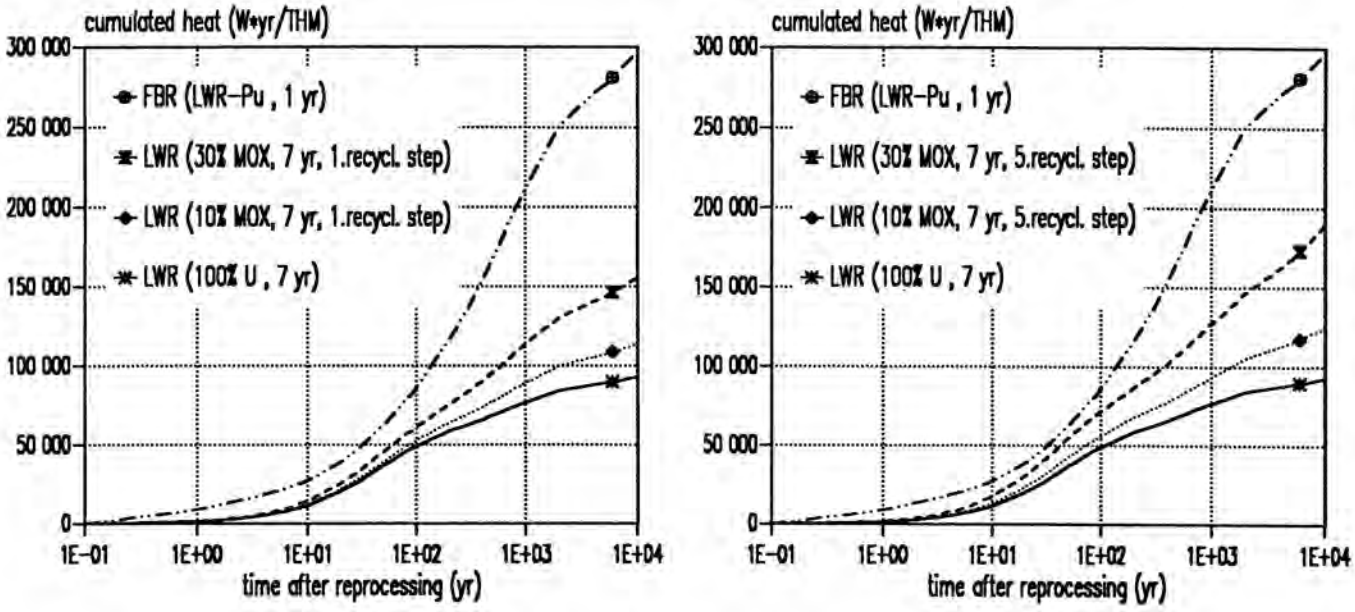
These curves are the lower and in most cases the upper limits of the thermal power released by HLWC from a common reprocessing of spent LWR uranium and LWR MOX fuels. An increasing MOX content (Fig. 1(a)) as well as spent fuels from higher Pu-recycling generations (Fig. 1(b)) increase the HLWC thermal power. For periods of about 6 to 20 years or longer than 3000 years the thermal power released by HLWC from LWR fuel with 30 % MOX content from the fifth Pu-recycling generation exceeds or is the same as in the case of HLWC from FBR fuel reprocessing.

Cumulated Heat

These differences in the thermal power are important for the thermal loading of a repository. A representative unit which gives a good overview for the interpretation of the results is the released or cumulated heat as a function of time after reprocessing. It is given in Fig. 2 and was calculated by time integration of the thermal power which was discussed above. These curves are representing the released heat in a repository with the prerequisite of a HLWC disposal immediately after reprocessing.

Fig. 2(a) demonstrates that vitrified HLWC from a common reprocessing of LWR uranium and 30 % MOX fuel (1. Pu-recycling step) causes by a factor of 1.7 more released heat as HLWC from uranium fuel reprocessing. To release the same heat after 10000 years of disposal, HLWC from fuel with 30 % MOX content needs an additional cooling time (interim storage) of about 100 years. This is more unfavourable in case of HLWC from MOX fuel of the fifth Pu-recycling generation and of HLWC from FBR fuel. Fig. 2(b) shows that for these types the heat release is increased by a factor of 2 and 3, respectively, compared with HLWC from U fuel.

The main heat contributions of the radionuclides after the period of 10000 years are listed in Fig. 3. This bar chart demonstrates for all types of HLWC a nearly uniform contribution of the fission products. Their main components are Sr/Y-90 and Cs/Ba-137. In the case of a HLWC from LWR uranium fuel the same amount is given by the actinides. This contribution significantly increases for HLWC from common reprocessing of LWR uranium and MOX fuel. It is mainly represented by Am-241, Am-243 and Cm-244. This figure additionally demonstrates that the multiple recycling of plutonium in LWRs causes more higher actinides (e.g. Am-243, Cm-244) in the HLWC. In the case of HLWC from FBR fuel, Am-241 yields the major contribution to the cumulated heat.



(a) 1. Pu-recycling step of LWR MOX fuel

(b) 5. Pu-recycling step of LWR MOX fuel

Fig. 2. Cumulated Heat of Vitrified HLWC From Reprocessed Spent LWR and MOX Fuels.

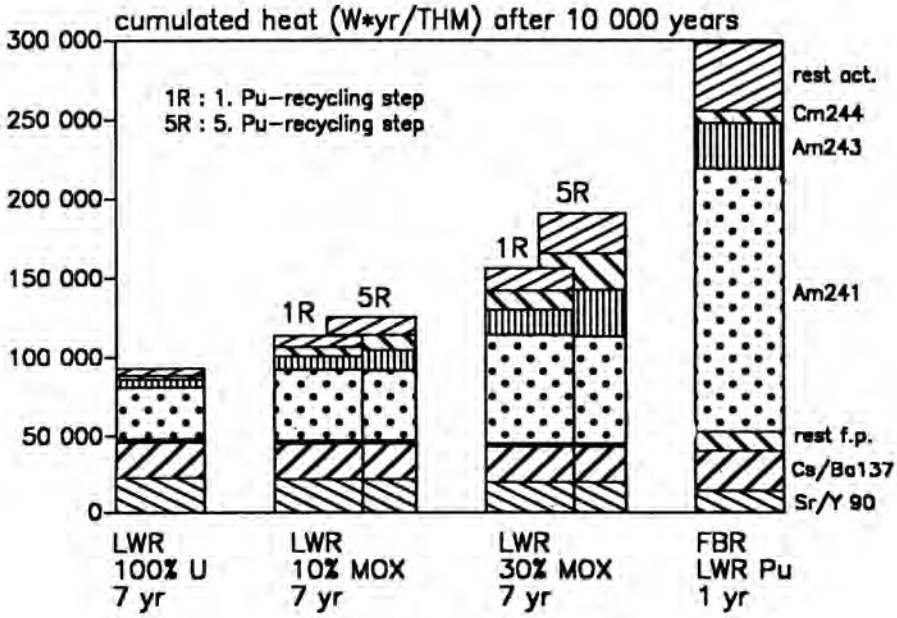


Fig. 3. Absolute Contributions of Radionuclides to the Cumulated Heat 10000 Years after Reprocessing.

Cooling Time of FBR Fuel

A reduction of the Am-241 content can be achieved by shortening the cooling time of the spent fuel. In the following reprocessing an earlier separation of the mother nuclide Pu-241 is achieved. Fig. 4 shows for a HLWC from reprocessed spent FBR fuel that the shortening of the cooling time reduces the Am-241 content and its contribution to the cumulated heat. Simultaneously, more short-lived fission products contribute to the heat and the total amount is slightly increased. The reduction of fission products for longer cooling times is compensated by a much bigger increase of Am-241. This demonstrates that a HLWC which results from reprocessing of a 1 year cooled spent FBR fuel effects a lower thermal loading of a repository than HLWC from longer cooled fuel.

SUMMARY AND CONCLUSIONS

Theoretical investigations have been performed in order to describe the thermal loading of a repository. Different types of HLWC from reprocessed spent reference-type LWR and FBR fuels with a burnup of 40 GWd/THM have been regarded. For the calculation of the thermal power and the cumulated heat (cumulated over a period of 10000 years) of vitrified HLWC the KORIGEN-code (3) has been used. The results can be summarized in the following way:

There are remarkable differences in the time dependant thermal power of HLWC from spent LWR uranium and LWR MOX fuels. An increasing Pu-fraction in the LWR fuel (increase in MOX content, increase in the Pu-recycling generation of the MOX fuel) leads to a higher thermal power of the HLWC. This growth in thermal power, especially for periods longer than 100 years causes a significant increase of the cumulated heat or the released heat in a disposal formation, respectively. After a period of 10000 years about 50 % of the cumulated heat of HLWC from LWR uranium fuel is caused by actinides. The growth of cumulated heat of HLWC with increasing Pu-content in the LWR fuel is exclusively effected by an increase of the actinide fraction. This results in a cumulated heat of a HLWC from reprocessed LWR fuel with 30 % MOX content (containing Pu from the fifth recycling generation) which is by a factor of about 2 higher as in case of HLWC from reprocessed LWR uranium fuel.

A vitrified HLWC from reprocessed spent FBR core and axial blanket material effects the biggest heat release. After 10000 years the cumulated heat of this HLWC type is by a factor of 3 higher than the cumulated heat of HLWC from LWR uranium fuel. The main fraction of about 80 % is caused by the actinides, especially Am-241. Furthermore, the calculations showed that the spent fuel cooling period

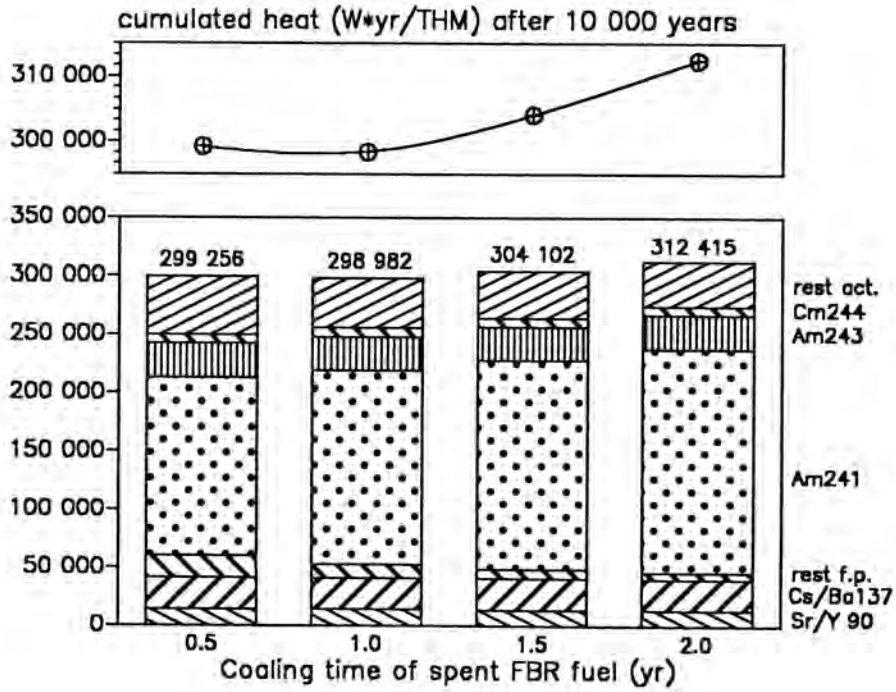


Fig. 4. Cumulated Heat of HLWC from Reprocessed Spent FBR Fuels for Different Cooling Times after Reactor Discharge.

of 1 year before reprocessing results in a lower cumulated heat than longer cooling periods.

The calculations demonstrated that the multiple recycling of plutonium in LWRs and the use of plutonium in FBRs significantly increases the thermal loading of the repository.

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