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Incineration of LWR-Plutonium (1. Generation and 2. Generation) in a Modular HTR using Thorium-based Fuel

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List of Content

1. Introduction

Large amounts of civil plutonium have accumulated in the world in burnt fuel from the operation of nuclear power reactors. Until the year 2000 they were estimated to amount to about 1400 Mg /1,2/. If plutonium is recycled in the form of U-Pu-MOX-fuel, separable second-generation Pu is produced. This can be avoided, if plutonium is burned in combination with thorium instead of uranium as fertile material.

The first section of this report presents fuelling strategies for

- burning as much plutonium (first generation) as possible per unit of produced energy
- minimizing the residual amount of this plutonium in the fuel for long term waste disposal

Only plutonium of the first generation – typical LWR-Pu - is regarded so far.

The second section reports recycling and incineration of second generation plutonium in a Pebble-bed HTR. Second generation plutonium means Pu-fuel, which already was recycled once in current LWRs and thus contains a distinctly lower fraction of fissile plutonium (about 40% - 50%) compared to plutonium of the 1. generation (about 70%).

2. Design of the Reactor and of the Fuel Element

The High Temperature Pebble-bed Reactor (HTR), which has been chosen as the basis for this study, is a close approximation of the HTR-MODUL-reactor, as it was designed in Germany by the INTERATOM company /3,4/. The small size of this reactor was required in order to avoid a release of fission products from the fuel elements by an inherent restriction of the fuel temperature to less than 1600 °C even in case of a complete loss of active cooling. The schematic of the reactor is illustrated in Fig. 1. The reactor core consists of a statistical filling of about 360000 pebble-shaped fuel elements, which are circulated about ten times through the core, until they reach their final burnup. It is surrounded by the graphite reflectors, which contain the control devices and the ducts for the supply of Helium, which is the cooling gas.

The research on Pu-burning has been based on the core design according to Fig.1 and Table I.

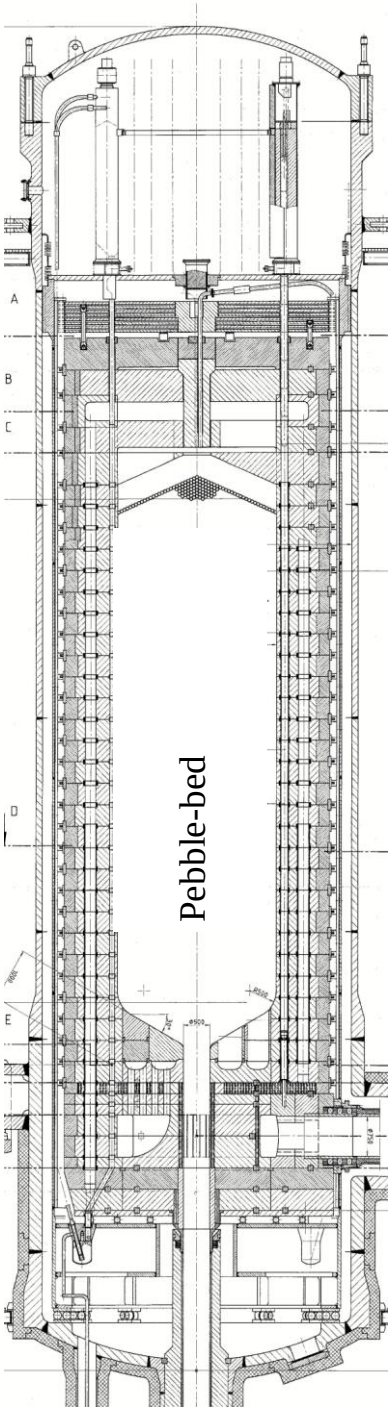

Fig. 1: Schematic of the HTR-MODUL-200 reactor layout

General Data

Thermal power	200 MW
Core height	9.43 m
Core diameter	3.0 m
Average power density	3.0 MW / m^3
Number of fuel elements/ m^3	5394

Fuel Element Specifications

Diameter of pebbles	6 cm
Diameter of fuel zone	5 cm
Density of graphite	1.75 g/cm^3

Specification of the coated particles:

Coating layers	C / C / SiC / C
Density	1.05/1.90/3.18/1.90 $g\ cm^{-3}$

Fuel	Diameter of Kernel	Maximum burnup
PuO_2	0.24 mm	800 MWd/kg
$(U\text{-}Th)O_2$	0.50 mm	120 MWd/kg

Assumed Isotope composition of Pu:

Fraction of	**$Pu^{238}/Pu^{239}/Pu^{240}/Pu^{241}/Pu^{242}$**
First generation Pu :	**0.01 / 0.62 / 0.24 / 0.08 / 0.05**
Second gener. Pu:	**0.05 / 0.36 / 0.35 / 0.10 / 0.14**

Table I: Specification of the HTR–MODUL Core for Plutonium burning

3. Methods and Computer Codes.

The numerical investigations within this study have been performed by means of V.S.O.P(99) /5/. This computer code system has been developed for the comprehensive numerical simulation of the physics of thermal reactors. It includes processing of cross sections, the setup of the reactor and of the fuel element, repeated neutron spectrum evaluation, neutron diffusion calculation in two or three dimensions, fuel burnup, fuel shuffling, reactor control and thermal hydraulics. The involved cross section libraries have been derived from the evaluated nuclear data files ENDF/B-V and JEF-1. The code can simulate the reactor operation from the initial core towards the equilibrium core.

4. Fuelling strategy

The HTR has some unique features, which make this system especially suitable to burn Pu on the basis of thorium-fuel:

- The fuel is loaded in the form of coated particles, which are embedded in the graphite matrix of the fuel elements. By this way, an exceptionally high heavy metal burnup can be achieved.
- The different fuel materials can be loaded into different types of coated particles. In a pebble-bed HTR the different fuel particles – Pu on the one hand and Th and U on the other hand – can also be loaded into different fuel elements. As they are continuously loaded and disloaded, these elements can even have different numbers of passes through the core – if desired – until they will reach their final burnup. On-line burnup measurements and the discrimination of LEU- vs. Th-fuel, respectively, have been successfully practiced for many years at the AVR reactor in Jülich (FRG) /6, 7, 8/, it was provided for the THTR-plant (FRG) /9/ and for the MODUL-200 HTR plant /10/.

The simultaneous use of different coated particle variants was originally developed for the high temperature reactors using prismatic fuel elements, as they were applied in the Fort. St. Vrain plant. Here, the fissile material was used in the form of small, so called "feed" particles reaching a burnup of about 90 % of its initial amount. Larger particles - "breed" particles- were made of ThO_2 .

In the present study the reference pebble-bed core design is fueled by use of a two-pebble-concept.
One type of pebbles (PU-FE) contains PuO_2 –coated particles with a diameter of 0.24 mm having a total of 3 g plutonium per pebble. The second pebble type (U/Th-FE) contains 20 g (HEU-Th)O_2 in the form of larger coated particles (diameter 0.5 mm). On

the one hand the addition of uranium to the thorium is necessary to sustain criticality – depending on the desired burnup of the fuel –, and, on the other hand, in order to achieve a prompt temperature increase of the resonance absorber, thorium, in case of an increase of the neutron flux, thus causing a prompt negative reactivity feedback. The uranium is highly enriched (93%) in order to minimize the build-up of Pu. While the (U-TH)-load of the fuel elements for the THTR-plant was 11g, values between 16 g and 20 g were provided in the frame of the German HHT-project (Hochtemperaturreactor mit Heliumturbine) and the reference maximum burnup was 120 GWd/t /11/. Elements containing up to 30 g heavy metal were developed within the same project /12/. A restriction to 20 g, however, should limit the fraction of coated particles, which will be damaged during the fabrication process of the elements, to 10^{-4} /13/.

As in particular the use of high-burnup plutonium particles cannot be regarded as proven technology, an irradiation program will be required to

- demonstrate that a burnup equal about 80 % "fissions per initial metal atom" (FIMA) can be achieved for the Pu (feed) particles without an inadmissible failure rate of the fuel coating,
- qualify (U-TH)-mixed oxide fuel elements loaded with 20g heavy metal,
- investigate the fission product retention of both the fuel element variants at a temperature level, which might occur in a loss-of-coolant accident.

A strategy for burning Pu can be optimized in view of two principal objectives. Today's main goal probably should be to reduce the separated amounts of Pu as soon as possible. This – in other words – means to maximize the amount of Pu depleted in nuclear reactors per unit of produced energy, which is equivalent to maximizing of the fractional power production by Pu in the reactors. Another important aspect, however, comes up with a view to intermediate storage of burned fuel and to final disposal of fuel without Pu-separation, as well as with respect to the non-proliferation aspect. From these points of view the minimization of residual Pu in the discharged fuel elements should be the main goal of the fuelling strategy. Here, the high burnup, which is achievable in case of HTR fuel elements, is a feature of particular importance. The positive features of this fuelling strategy, of course, imply the need to handle highly enriched uranium.

5. Results for 1. Generation Plutonium

5.1 Mass Balance of the Fuel

Table II shows a comparison of two fuelling strategies for the incineration of spent LWR-Pu in the considered HTR core. The first strategy is designed to achieve a high Pu burning ratio, the second one to achieve an especially small amount of residual Pu in the discharged fuel elements. Table III displays the corresponding mass balance of the plutonium and of the fissile uranium.

Both cases apply two kinds of fuel elements, as it has been described above. About half the reactor power is produced by fissions of the Pu. The charged Pu is depleted by 81 % (Table III, case b) and about 500 kg Pu are incinerated per GWa of produced electrical energy, assuming the efficiency 0.4 for the HTR power plant. In case a) the burnup period of the fuel elements is reduced from 11 down to 7.3 years of full power, and thus the average burnup of the fuel is lowered to a standard operation value of the German AVR reactor. In consequence the amount of Pu burned per GWa_{el} increases by 30 %. On the other hand, the residual Pu of the discharged fuel also increases from 19 % to 31 % of the initial amount. The requirement of uranium is similar in both cases.

An outline of the isotopic composition of the discharged plutonium and of the discharged uranium is given in Table IV for "Case a". The bred U^{233} and the remaining amount of U235 are denatured by U^{234} – produced by neutron capture of U^{233} -, by U^{236} –produced by neutron capture of U^{235} –and by U^{238}. The fraction of U^{233} is only 30%. So, in view of the proliferation aspect, the question obviously is not, whether to accept the production of U^{233}, but whether and where – in the sense of the non-proliferation treaty- to accept the handling of HEU in order to incinerate the plutonium.

	a) High Pu-burning ratio		b) Low residual Pu in disch. Fuel	
	Pu-FE (50%)	U/Th-FE (50%)	Pu-FE (50%)	U/Th-FE (50%)
Pu	3 g	---	3 g	---
Th	---	18 g	---	16.7 g
U (HEU)	---	2 g	---	3.3 g
Incore Time	7.3 F. P. Years		11.0 F. P. Years	
Fractional Power Prod.	65 %	35 %	52 %	48 %
HM-burnup	595 MWd/kg	58 MWd/kg	700 MWd/kg	116 MWd/kg
Average	128 MWd/kg		192 MWd/kg	

Tab. II: Fuelling Strategy of the HTR-MODUL Reactor for Burning LWR – Pu

	a) High Pu-burning ratio		b) Low residual Pu in disch. fuel	
	Pu-FE (50%)	U/Th-FE (50%)	Pu-FE (50%)	/Th-FE (50%)
Pu charged	929 kg/GWa$_{el}$	----	615 kg/GWa$_{el}$	---
Pu dischgd.	265 kg/GWa$_{el}$	23	93 kg/Gwa$_{el}$	26
Pu burned	664 kg/GWa$_{el}$ Σ 641	- 23	522 kg/GWa$_{el}$ Σ 496	- 26
Pu burned/ Pu charged	0.69		0.81	
U^{235} charged	------ kg/GWa$_{el}$	578	------- kg/GWa$_{el}$	624
U^{235} discharged	------ kg/GWa$_{el}$	251	------- kg/GWa$_{el}$	171
U^{233} produced	------ kg/GWa$_{el}$	161	------- kg/GWa$_{el}$	116

Tab. III: <u>Mass Balances</u> for the HTR-MODUL burning LWR-Pu

	Pu-FE	U/Th-FE
U3/U4/U5/U6/U8 (%)	----------	30/ 3/48/13/ 6
Pu8/Pu9/Pu0/Pu1/Pu2 (%)	0/13/20/38/29	93/ 7/ 0/ 0/ 0

Table IV: Isotopic Fraction of U and Pu in burnt Pu-Fuel Elements and in burnt HEU/TH-Fuel Elements (Case a)

5.2 Temperature Coefficients

Parametric study on the temperature coefficients of a HTR for Pu-burning showed the need for a relatively large Pu-load of the fuel elements, favorably about 3 g Pu . The result is a "hard" thermal neutron spectrum, which favors the parasitic absorption of neutrons in the resonance of the Pu^{240}– absorption cross section at the energy 1 eV. Its increase with the moderator temperature dominates some others –partly contrary- spectral effects. Thus, the value of the moderator coefficient is strongly influenced by the fraction of Pu^{240} in the fuel.

The temperature coefficients of the regarded reactor are listed in Table VI for the case Aiming at the high Pu-burning rate (case a). They were calculated for full-power operation of the core. Increasing the operation temperature of the fuel and of the moderator, respectively, at each local position by the same value simultaneously, results in the reactivity change described by the given coefficients. All temperature coefficients are negative and sufficiently large in the absolute value.

Doppler coefficient	**$-1.71 \cdot 10^{-5}$ / K**
Moderator coefficient	**$-4.46 \cdot 10^{-5}$ / K**
Total Temp.- coefficient	**$-6.17 \cdot 10^{-5}$ / K**

Table VI: Temperature Coefficients ($\Delta K_{eff} / \Delta T$) at operating temperature (Design for high Pu – burning ratio)

5.3 Summary and Conclusions, 1. Generation Plutonium

Within the scope of global efforts to reduce the large stockpiles of plutonium by making use of this fissile material for the production of electrical energy, one goal is to avoid the production of second-generation plutonium. This objective should lead from today's use of U-Pu-MOX-fuel to burning plutonium in combination with thorium as fertile material. The coated –particle fuel of HTRs allows an incineration fraction of the initially loaded by about 70 % during the lifetime of the fuel elements, using typical LWR-Pu.

In view of a more rapid reduction of the existing Pu-stockpiles the burnup of the fuel may be lowered in order to increase the quantity of plutonium, which is incinerated per unit of produced electrical energy. A reduction of the average discharge burnup of the fuel from 190 MWd/ kg to about 130 MWd/ kg, e.g. increases the incineration rate of the plutonium from about 560 kg to about 640 kg per GWa_{el} on the one hand, but on the other hand increases the fraction of residual plutonium from roughly 20 % to 30 % of the initial amount.
The temperature coefficients of the reactor –both the Doppler and the moderator coefficient- are sufficiently negative over the whole applied temperature range of reactor operation .

6. Results and Conclusions for 2. Generation Plutonium

Three basic fuelling strategies for 2. Generation Pu have been investigated and compared:

1. The use of the plutonium together with a basic fuel, which is thorium-uranium mixed oxide, which also is the reference strategy of ISR for the incineration of first-generation plutonium (see section 5), and – as possible alternatives-,
2. The use of pure plutonium fuel
3. The use of plutonium together with 20 % enriched Uranium.

The most important results of this study are summarized in Table VII.

Table VII: Comparison of different fuelling strategies for the incineration of ‚Second-generation' Plutonium in HTRs

Fuel elements	Heavy metal.-burnup (MWd/to)	Average burnup (MWd/to)	Pu charged (kg/GWa-el)	Plutonium burned (kg/GWa-el)	Ratio Pu burned/ Pu-charged (%)
50% Pu, Typ1 50% Th, Typ1	522 000 122 000	174 000	683	450	66
50% Pu, Typ1 50% Th, Typ2	495 000 112 000	200 500	1048	643	61
100% Pu, Typ2	428 000	---	2050	1020	50
100% Pu, Typ3	416 000	---	2093	968	46
50% Pu, Typ1 50% U	145 000 30 000	55 000	3725	503	14

Fuel element types:

Pu, Type 1: 3g Plutonium 2. Generation / fuel element

Pu, Type 2: 1g Plutonium 2. Generation / fuel element

Pu, Type 3: 0.5g Plutonium 2. Generation / fuel element

Th, Type 1: 20g (Th + HEU)-MOX / fuel element

Th, Type 2: 10g (Th + HEU)-MOX / fuel element

U: 10g U, (20% ^{235}U) / fuel element

Generally, the incineration of 2.-generation plutonium is much less effective than as it turned out to be for 1.-generation plutonium. The reason for this is the much larger degree of denaturing of the fissile plutonium isotopes by the isotopes ^{238}Pu, ^{240}Pu and ^{242}Pu, which are not fissioned by thermal neutrons. Compared to what resulted for 1.-generation Pu, the amount of thorium (and Uranium) loaded to the driver fuel should be reduced by about 50% in case of 2.-generation („Th, Typ2“ instead of „Th, Typ1“, see Tab. VII). By this mean, the amount of plutonium, which is incinerated per year, is increased by 43 %. The relative fraction of incinerated Pu (burned / charged), however, is slightly reduced (from 66% to 61%), simultaneously.

Changing over to loading the reactor with exclusively plutonium-fuel elements requires a strong reduction of the amount of Pu per fuel element (from 3g to 1g Pu / fuel element, “Pu-type 2”) in order to achieve a critical core. The amount of Pu burned per GWa_{el} then increases to more than 1000 kg, the fraction of residual plutonium in the discharged fuel elements, however, is also increased to about 50%, simultaneously.
It should be noted, that further R&D-work, concerning the dynamic features of such a reactor will be required in the future. In particular, more investigations will have to make clear, whether or not inherently safe, i.e. stable properties of the reactor dynamics will be maintained in spite of the loss of the doppler-effect of the breeding material (Th or U) and of the combined prompt, negative feedback of the fuel temperature to the reactivity of the system.

Pu-incineration by means of a use of low-enriched uranium as basic fuel turns out to be the by far most unfavorable variant of the regarded fuelling strategies. Here, the production of new plutonium by the breeding effect of ^{238}U is nearly as big as the destruction rate by fissions. As consequence, the amount of plutonium in the unloaded fuel elements is lower by only 14% compared to the start of the irradiation.

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