RECENT NEUTRON PHYSICS INVESTIGATIONS FOR THE BACK END OF THE NUCLEAR FUEL CYCLE

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ABSTRACT

The neutronic calculations for the back end of the nuclear fuel cycle at the Forschungszentrum Karlsruhe (FZK) cover 3 main areas. The transurania buildup in the widely used Pressurized Water Reactors (PWR) is investigated for scenarios with and without spent fuel reprocessing. The investigations are based on calculational tools originally developed for the design of tight lattice light water reactors. These methods are qualified for fast, epithermal and thermal reactor systems. For the consumption of multi-recycled plutonium in PWRs, the use of enriched uranium is proposed. The investigations of the incineration potential of transurania and fission product isotopes with fast reactor and accelerator-driven systems are more futuristic compared to the investigations for the PWR. The fast reactor investigations are the redirection of traditional FZK activities in this area. The work now concentrates on nuclear waste transmutation with special design fast spectrum reactor cores and is embedded within the framework of the European CAPRA project. The investigations for accelerator-driven systems (ADS) started some years ago with the implementation and the qualification of the required calculational tools. The work is partially supported by the IABAT program of the European Community. First results for the incineration of plutonium with ADS are presented.

1. Introduction

One of the most challenging problems at the end of this century, from the technical, ecological and political point of view, is the back end of the nuclear fuel cycle. Besides the existing considerable amounts of transurania and fission products from military and commercial applications of nuclear fission, nuclear power plants all over the world are producing nuclear waste, containing large amounts of potential new nuclear fuel. In Germany the Atomic Act prescribed for a long period of time the recycling of spent reactor fuel for the use for further energy production. However, a recent amendment now also allows the direct disposal of spent fuel.

An important objective of the R&D program at the Forschungszentrum Karlsruhe (FZK) consists in maintaining and preserving existing capabilities to judge the potential of the various alternative options for the back end of the nuclear fuel cycle. The present contribution is restricted to recent developments in the neutron physics area. These investigations focus on a qualified assessment of the capabilities of a number of different concepts for the incineration of transurania and fission product isotopes in comparison to direct disposal of spent fuel.

The following topics will be discussed:

- Transurania buildup in modern pressurized water reactors (PWR), including plutonium multi-recycling.
- Plutonium incineration in fast reactors.
- Developments and application of methods for the investigation of accelerator driven systems (ADS).

The recent investigations for actual reactors concentrate on the reduction of buildup of transurania in **PWRs**, being currently the most utilized nuclear reactor type. These investigations are a continuation of the activities for tight lattice light water reactors, aiming to preserve uranium fuel resources by enhanced plutonium generation. The applied methods are qualified for fast, epithermal and thermal reactor systems. In the present investigations it is shown, that the use of enriched uranium in the MOX fuel during plutonium multi-recycling in PWRs, may lead to a nearly constant plutonium inventory after about 60 to 100 years. The amount of plutonium at that time is about half the quantity of plutonium to be produced without fuel reprocessing.

The **fast reactor** investigations are based on the comprehensive activities in the area of fast breeder reactors at the Forschungszentrum Karlsruhe. Within the European CAPRA program the aim is now to incinerate plutonium in future reactors with hard neutron spectra. As an extreme case, the use of uranium-free fuel is considered.

For the **accelerator driven systems** (ADS) calculational procedures have been made available and validated, especially by benchmark participation. The application of alternative methods for transport calculations has been studied in some detail. Recently first results for plutonium incineration have been obtained.

2. Investigations for Pressurized Water Reactors.

The use of $(UPu)O_2$ mixoxide (MOX) fuel in PWRs is proven technology in several countries, e.g. France and Germany. Up to about 30% of the fuel assemblies (FA) in existing PWR cores are replaced by MOX instead of UOX assemblies. A main objective during this replacement is to maintain the safety characteristics of the PWRs, especially the coolant density reactivity coefficient (CDRC) and the fuel Doppler coefficient (DC). From earlier studies, e.g. in reference [1], it is well-known that high plutonium fractions in the MOX fuel of PWRs may lead to problems with the CDRC. Due to the neutron captures in fissile isotopes the fraction of non-fissile isotopes in the plutonium increases if its irradiation time is increased, e.g. by enlargement of the burnup or by multi-recycling of the spent fuel. As a consequence, the plutonium fissile (and also the total) fraction of MOX fuel with depleted or natural uranium must be increased considerably during plutonium multi-recycling and one must take care concerning the CDRC. In a recent study [2, 3] the buildup of transurania in PWRs has been investigated in some detail, both for direct disposal of spent fuel from UOX cores and for plutonium multi-recycling. In the present paper some results for the transurania buildup in UOX cores and in pools of PWRs with a mixing of UOX and

MOX cores will be presented. All calculations are based on a reactor lattice from benchmark investigations on plutonium multi-recycling, which were performed in collaboration of Forschungszentrum Karlsruhe (FZK) and Electricité de France (EDF) [4, 5].

2.1. **Transurania buildup in UOX fuel.** In table 1 a summary is given of the transurania buildup in modern PWRs with UOX fuel in dependence of the U^{235} enrichment for discharge burnups of 33 and 50 gigawattdays pro tonne heavy metal (GWD/THM). The weights are normalized to 1 tonne initial heavy metal (TIHM). The burnup calculations are performed on the basis of the realistic power rating of 164 W/cm, corresponding to ≈35 W/g. One tonne of spent fuel from modern PWRs contains also about 10..13 kg of transurania isotopes, mainly plutonium. These PWRs have a loading of about 80 tonnes of heavy metal fuel per gigawatt electric (GWe), leading to about 800..1000 kg transurania per GWe per reactor core life or to amounts of around 200 kg discharged plutonium per GWe.a energy production.

Table 2 shows the influence of discharge burnup and U^{235} enrichment on the plutonium composition. The fissile fraction decreases significantly if the discharge burnup is increased from 33 to 50 GWD/THM. Higher U^{235} enrichment causes less in situ plutonium burning and leads to higher fissile fractions of the plutonium.

2.2. **Plutonium multi-recycling in PWRs.** The use of fuel assemblies with MOX fuel is proven technology in PWRs, e.g. in France and in Germany. Until now mainly good quality plutonium with high fissile fractions has been utilized for discharge burnups of ≈35 GWD/THM. The potential of plutonium multi-recycling in PWRs has been analyzed in some detail in a common benchmark investigation of FZK and EDF [4, 5]. These investigations were based on simplified lattice calculations. The main result was, that after a small number of recyclings the fissile fraction of the plutonium becomes so unfavourable, that the fissile enrichment of the plutonium has to exceed 6..7% if about 50 GWD/THM discharge burnup should be reached with natural or depleted uranium in the MOX. With such plutonium fractions in the MOX fuel it cannot be guaranteed, that the CDRC keeps satisfactory. Starting from this experience, in reference [2] whole core calculations for pools of PWRs with full UOX and full MOX cores have been performed. In figure 1 the priciple of this reactor scenario is shown. To avoid problems with the CDRC, the fissile fraction of the plutonium was restricted to ≈6%. In order to get enough excess reactivity at reactor startup to obtain the required discharge burnups, U^{235} enriched uranium is used if necessary. In table 3 important results are summarized for the near equilibrium situation. For discharge burnups of 33, 40 and 50 GWD/THM the normalized plutonium production in UOX cores and the net plutonium incineration in the MOX cores are given. The ratio of these plutonium masses is nearly independent of the discharge burnup:≈0.6. This means, that the plutonium inventory is nearly constant in a pool of PWRs with a ratio of 3 MOX to 5 UOX cores. In table 3 we also may observe, that the required U^{235} enrichment increases rapidly with the increase of the discharge burnup if the fissile plutonium fraction of the MOX fuel is restricted to 6%. In figure 2 the normalized buildup of plutonium for a period of about 100 years is shown. The calculations are based on ex-core times of 7 years (cooling + reprocessing) and 3 years fabrication time. For each discharge burnup a suitable

number of reactor burnup cycles during these 10 years ex-core time is applied. We may observe that after 80 to 100 years the plutonium inventory is nearly constant at a level of about half the value of the case without plutonium multi-recycling.

3. Investigations for Fast Reactors.

Fast reactors have the well-known advantages of

- low values of $\alpha = \frac{\sigma_c}{\sigma_f}$ for most of the heavy isotopes leading to high values of $\eta = \frac{\nu\sigma_f}{\sigma_a}$ and a resulting high neutron excess which, in former times, was used to breed fresh fissile material and
- low mean cross section values for most of the fission products so that even for fairly high burnups the neutron balance is not too much deteriorated by neutron absorptions in these fission products.

On the other hand, fast reactors suffer from the disadvantages of

- generally rather small average cross sections requiring high concentrations of fissile material and a rather high neutron flux intensity and
- their fairly large fraction of high energy neutrons (E$\geq$0.1MeV) which are inducing pronounced radiation damage in structural materials.

These pecularities of fast reactors with "hard" neutron spectra are responsible for the limitation of the residence time of fuel assemblies which can withstand damage rates of about 100-200 displacements per atom (DPAs). The corresponding neutron fluences of several $10^{23} n/cm^2$ are usually not sufficient for burning almost completely the initial fissile content of the fuel. Furthermore, due to the conversion of fertile to fissile material, the burnt fuel still contains a large fraction of fissile (and fertile) isotopes so that reprocessing is practically mandatory for fuel irradiated in fast reactors.

The incineration capabilities of fast reactors with conventional oxide (MOX) fuel is essentially limited by the solubility of the MOX fuel which becomes much worse when the Pu-content is increased above about 45% (as is well known, there exists a rather strong correlation between the fissile enrichment and the maximum incineration rate which can be achieved). Accepting the above enrichment constraint, an incineration rate of roughly 70 kg(Pu)/TWh(e)$\approx$600 kg(Pu)/GWa(e) was obtained for a CAPRA-type reactor with MOX fuel. Of course such a burner reactor has no longer any fertile blankets as previously encountered in fast breeder reactor designs. Moreover, so-called diluents were loaded in the core to increase the neutron leakage so that a large fast reactor with a power of 1500 MW(e) could be loaded with fuel having a significantly larger Pu-content than earlier breeder-related designs such as SUPERPHENIX or EFR. In addition to burning plutonium, special investigations were devoted to the transmutation of minor actinides (MAs). The addition of MAs (especially of Np^{237}) causes a moderate reduction of the burnup reactivity swing but causes a pronounced reduction of the Doppler effect due to the large absorption cross

sections of these MAs in that part of the resonance region which mainly contributes to the Doppler effect. This drawback of MA addition can be mitigated or compensated in such a way that the ratio of coolant void effect to Doppler effect is similar to that of a burner reactor without MA addition by replacing diluent or inert material in diluent subassemblies (S/As) or in so-called "empty" pins of fuel S/As by a suitable moderator material like BeO or $B_4^{11}C$ (boron carbide with a practically vanishing content of B^{10}). This enables a fast reactor to incinerate at least its self-generated MAs with a non-negligible capacity to accept an additional amount of MAs e.g. originating from LWRs. Having in mind the complications and economic aspects of fabricating and handling pins and S/As loaded with pellets containing significant amounts of americium it seemed appropriate to concentrate such highly radioactive MAs in special S/As, placed in reflector positions of the core. Obviously this leads to only a small degradation of core performance parameters (e.g. safety related reactivity coefficients) by the addition of MAs (in the reflector) and an operational behaviour of the MA-loaded reflector S/As which is similar to that of conventional radial breeders S/As.

As a particular design of an extraordinary burner reactor a so-called U-free core was studied. In order to comply with solubility requirements, nitride fuel was chosen and the necessary Doppler feedback was delivered by using a fairly "dirty" Pu isotopic composition with high fractions of the even isotopes Pu^{240} and Pu^{242}. This version of a burner could be considered as adequate to deal with multi-recycled LWR-Pu and, therefore, possibly as representative for the later or last phase of nuclear energy production. It would allow to come close to the theoretical limit of about
120 kg(Pu)/TWh(e)≈1000 kg(Pu)/GWa(e).

4. Investigations for Accelator-Driven Systems.

ADS investigations started at FZK about 5 years ago with the installation of parts of the HERMES[6] code version of KFA Jülich. The module HETC for the calculation of intermediate energy processes and the Monte Carlo module MORSE for transport calculations below 20 MeV are operational both on IBM-mainframe computer and on UNIX-workstations. Moreover interfaces were established between HETC, the Monte Carlo program MCNP[8] and the discrete ordinates transport code TWODANT[7]. For burnup investigations the program PROSDOR[9] was developed. It consists of a semi-automatic sequence of the codes HETC, MCNP-3A and the depletion code KORIGEN[10]. KORIGEN was developed at FZK from the original ORIGEN code from Oak-Ridge. Some special provisions for the PROSDOR application were made. Recently for the burnup calculations a sequence of HETC, MCNP/TWODANT and the burnup code KARBUS[1] has been established. KARBUS was developed at FZK for more general fission reactor burnup calculations. Its depletion part is also based on ORIGEN formalisms. Much effort has been spent for best estimate determination of one-group cross sections.

4.1. **Validation work.** The first activity after installation of part of the HERMES package was to gain experience with the application of the code. This could be realized

by a successful participation to the NEA/NSC international code comparison benchmark for intermediate energy nuclear data for thick targets [11]. Generally the FZK results agree satisfactorily with the other solutions. Further extensive comparisons were performed between the Monte Carlo code MCNP and the discrete ordinates code TWODANT [12] with the objective to replace the time-consuming burnup sequence PROSDOR/KORIGEN by the faster sequence HETC/TWODANT/KARBUS. These comparisons are based on non-multiplicative targets and slightly subcritical systems with thorium fuel. As an example, in table 4 one-group data for U^{233} from MCNP and TWODANT calculations for a Th-U^{233} ADS are compared. With exception of the threshold cross sections (n,2n) and inelastic scattering, the differences between the calculational methods are small. These threshold processes must be analyzed carefully because of their sensitivity to spectral changes in the results of the calculations. Especially the (n,2n) cross sections may influence significantly the buildup of actinide isotopes, resulting from such processes.

4.2. **Applications.** First results have been published by Segev et. al. [13] for different applications, mainly transmutation of long-lived minor actinides and fission products. Recently investigations on incineration of LWR-plutonium have been started. Figure 3 shows preliminary results for the time dependence of the isotopic concentrations in a Th-Pu ADS with 3000 MWth. The agreement between the MCNP and TWODANT results is very satisfactory. We may observe that the incineration of the plutonium is counterbalanced by the buildup of U^{233}. To judge such a system it will be necessary to handle this U^{233} in a proper way. The K_{eff} of this system varies from $\approx$0.98 to $\approx$0.96. The corresponding proton currents for achieving 3000 MWth are in the range of 18 to 40 mA. Total voiding of the ADS leads to small negative ΔK_{eff} changes. The hot to cold reactivity change is less than +1% in K_{eff}, so the ADS remains safely subcritical during shutdown.

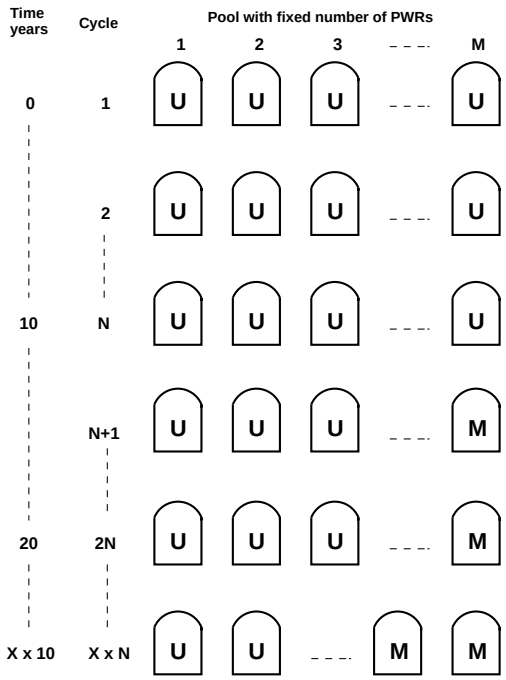

Figure 1: Scenario for plutonium multi-recycling in a pool of PWRs.

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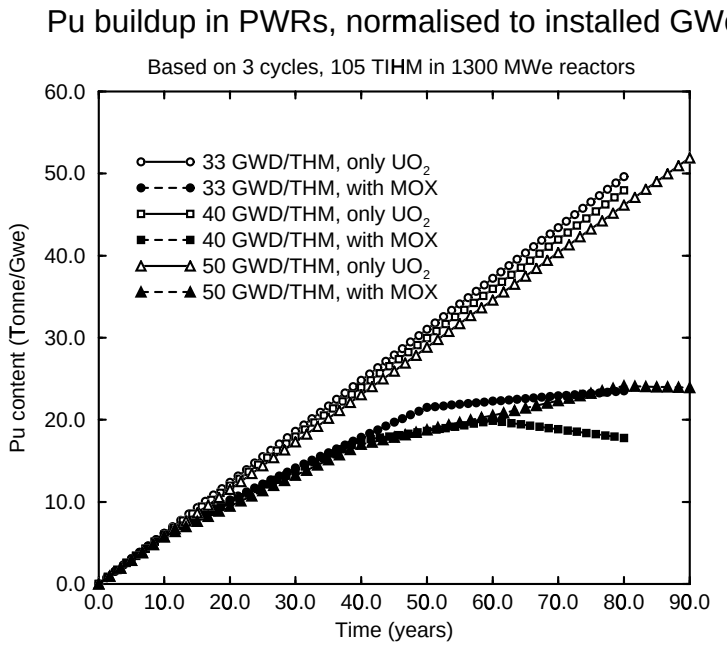

Figure 2: Plutonium buildup in PWRs for burnups of 33, 40 and 50 GWD/THM.

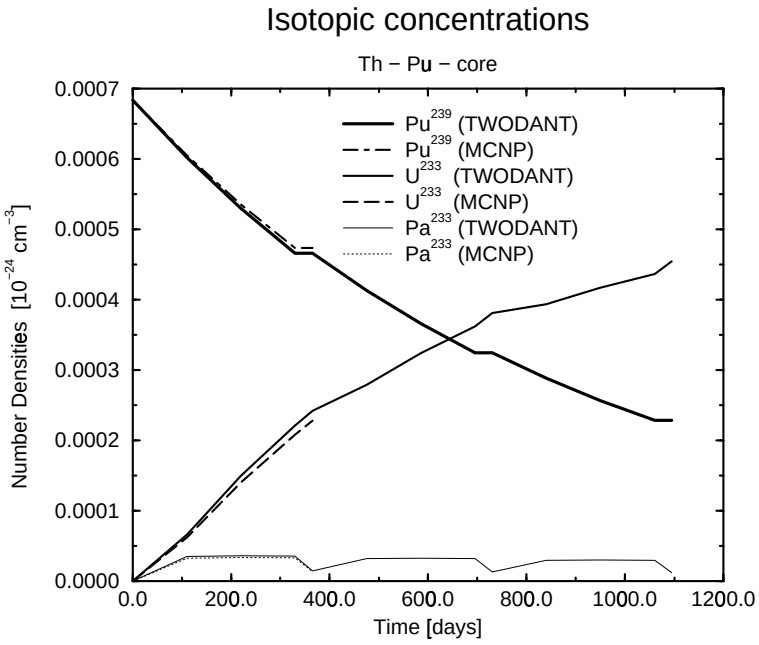

Figure 3: Th-Pu ADS, time dependence of isotopic composition.

Material (kg/TIHM)	Burnup							
	33 GWD/THM				50 GWD/THM			
	U^{235} Enrichment				U^{235} Enrichment			
	3.2%	3.5%	4.0%	4.5%	3.2%	3.5%	4.0%	4.5%
Pu	9.680	9.666	9.640	9.612	11.833	11.874	11.946	12.017
Np^{237}	0.4254	0.4306	0.4368	0.4408	0.6453	0.6650	0.6916	0.7119
Am^{241}	0.0370	0.0373	0.0374	0.0371	0.0540	0.0565	0.0604	0.0638
Am^{243}	0.1002	0.0869	0.0694	0.0561	0.3220	0.2914	0.2467	0.2095
Transurania	10.243	10.220	10.184	10.146	12.855	12.886	12.944	13.003

Table 1: Transurania buildup, power rating 164 W/cm

U^{235} (%)	Burnup											
	33 GWD/THM						50 GWD/THM					
	Pu isotope (%)					Fiss. (%)	Pu isotope (%)					Fiss. (%)
	238	239	240	241	242		238	239	240	241	242	
3.2	1.5	56.8	22.1	14.0	5.6	70.8	2.9	47.6	24.5	14.8	10.2	62.4
3.5	1.5	58.3	21.3	13.9	5.0	72.2	2.9	48.8	24.0	14.9	9.4	63.7
4.0	1.4	60.6	20.3	13.5	4.2	74.1	2.8	50.8	23.2	15.0	8.2	65.8
4.5	1.3	62.8	19.1	13.1	3.7	75.9	2.7	52.8	22.3	15.0	7.2	67.8

Table 2: Plutonium compositions, power rating 164 W/cm

Target-burnup (GWD/THM)	Pu-balance (kg/TIHM)		Ratio UOX / MOX	U^{235} enr. (%)
	UOX	MOX		
33	+9.6	-16.2	0.6	0.7..1.0
40	+10.6	-18.5	0.6	2.0..2.5
50	+11.9	-21.0	0.6	3.5..4.0

Table 3: Near equilibrium cycles in pools of PWRs with UOX and MOX fuel.

	TWODANT 69 groups		MCNP-4A	MCNP-3A	$\frac{TW-MC}{MC}\%$ [1]
	k_{eff}	inhomogen. source	k_{eff}	inhomogen. source	inhomogen. source
σ_{tot} $[\frac{1}{cm}]$	9.5640	9.5738	10.0466 (.0016)	10.0793 (.0019)	-5.0
σ_{el}	6.5330	6.5403	7.0542 (.0017)	7.0528 (.0019)	-7.3
$\sigma_{(n,2n)}$	1.5172E-3	2.1227E-3	7.9018E-4 (.0297)	1.6015E-3 (.0421)	32.5
σ_{fiss}	2.2597	2.2622	2.4602 (.0017)	2.4631 (.0208)	-8.2
σ_{inel}	0.5852 [2]	0.5841 [2]	0.3176 (.0017) [3]	0.3143 (.0461) [3]	85.9
$\sigma_{(n,\gamma)}$	0.1847	0.1850	0.2137 (.0021)	0.2141 (.0208)	-13.6
ν $[\frac{neutrons}{fission}]$	2.5393	2.5406	2.5240 (.0014)	2.5266 (.0208)	-0.56
k_{eff}	0.9035	0.9142 [4]	0.9183 (.0016)	0.9510 [4] (.0204)	

[1] TW=TWODANT MC=MCNP

[2] inelastic from discrete levels and from energy continuum

[3] only continuum inelastic

[4] determination of k_{eff} - values for calculations with external neutron source with formula from reference [8]:

$$k_{eff} = \frac{M - R^{(0)}(\sigma_{n,xn}) - 1}{M - \frac{1}{\nu}}$$

with $M = R(\nu\sigma_{fiss}) - R(\sigma_{fiss}) + R(\sigma_{n,xn}) + 1$

where R is the corresponding reaction rate.

$R^{(0)}$ means reaction rate from criticality calculation

Table 4: U-Th ADS, comparison of one-group data of U^{233}