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Investigations related to the buildup of transurania in pressurized water reactors

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Abstract.

The backend of the nuclear fuel cycle is of large importance for the near future. The German atomic act now allows both the direct disposal and the reprocessing of the spent fuel from nuclear reactors. Nowadays most of the running nuclear reactors in the world are based on light water technology with a preference for pressurized water reactors (PWR). In the present study systematic investigations related to the buildup of transurania in PWRs are described.

For a scenario without reprocessing, parametric variations of the discharge burnup and of the U^{235} enrichment have been performed. Generally, an increase of the discharge burnup in PWRs, due to the in-situ burning of plutonium, leads to a decrease of the specific plutonium production and to an increase of the neptunium and especially of the americium buildup. Higher U^{235} enrichments act against these trends. The fissile content of the plutonium decreases with increasing burnup.

From the viewpoint of saving energy resources, it is advantageous to apply reprocessing of the spent fuel. Plutonium recycling in PWRs is already a proven technology, e.g. in France and in Germany. In these applications PWR cores partly are fueled with uranium/plutonium mixed oxide (MOX) fuel assemblies. The equivalency of uranium oxide (UOX) and MOX fuel assemblies in the same PWR core have lead to some problems in the past. Based on experiences in French and German PWRs, a specification for the fissile fraction of the MOX fuel has been derived. The problem of plutonium multi-recycling in PWRs has been studied with the help of whole core calculations for full MOX cores with appropriate scenarios for the mass flows. These whole core calculations need fissile plutonium fractions in accordance with the results of the investigations for the equivalency of UOX and MOX fuel assemblies, mentioned before. The analysis of the coolant density reactivity coefficient indicates that the plutonium fissile fraction should be limited to $\approx$6%, if safety related parameters have to be maintained. The long term investigations are based on a scenario with pools of PWRs, consisting of only UOX cores at the beginning. As soon as enough plutonium is produced in this pool, UOX cores are replaced by full MOX cores. For the ex-core periods, 7 years of cooling and reprocessing time and 3 years of fabrication time are chosen. The plutonium for the next cycle is obtained by mixing all available plutonium from the UOX and the MOX cores. The fissile fraction of the plutonium is limited to $\approx$6%; if necessary, enriched U^{235} is used to meet criticality conditions. This scenario for plutonium multi-recycling leads to a near to equilibrium plutonium composition with constant inventory after about 60 to 80 years. At this time the amount of plutonium is about half the value, compared to a scenario without plutonium recycling. This means that large savings of plutonium buildup may be obtained by plutonium recycling in PWRs. The buildup of neptunium amounts to 5..6% of the plutonium in one cycle and is not sensitive to plutonium recycling. However, the buildup of americium reaches the same order of magnitude as the neptunium if plutonium recycling is applied. Otherwise the amounts of americium are significantly smaller than those of neptunium.

In a closed fuel cycle with plutonium recycling also large amounts of recycled uranium (RU) are produced. Preliminary calculations indicate that this RU also may be used in PWRs. The enrichment of this RU may be realized directly in an enrichment plant for RU or by blending with small amounts of highly enriched fresh uranium.

Untersuchungen zum Aufbau von Transuranen in Druckwasserreaktoren. Kurzfassung.

Die Entsorgung der nuklearen Abfälle ist eines der wichtigen Probleme für die nahe Zukunft. Das deutsche Atomgesetz in seiner aktuellen Fassung erlaubt sowohl die Wiederaufbereitung als auch die direkte Endlagerung von abgebrannten Brennelementen (BE). Die meisten in Betrieb befindlichen Kernreaktoren auf der Welt beruhen auf der Leichtwasser Technologie, mit einer Bevorzugung von Druckwasserreaktoren (DWR). Die vorliegende Arbeit beschreibt eine Reihe von systematischen Untersuchungen zum Aufbau von Transuranen in DWR.

Für ein Szenario ohne Wiederaufbereitung wurde der Aufbau von Transuranen als Funktion des Entladeabbrands und der Anfangsanreicherung des Urans parametrisch untersucht. Im allgemeinen gilt, daß - wegen der in-situ Verbrennung des Plutoniums im Urankern - eine Erhöhung des Entladeabbrands zu einer Verringerung des spezifischen Plutoniumaufbaus und zu einer erhöhten Erzeugung von Neptunium und insbesondere Americium führt. Eine Erhöhung der Urananreicherung wirkt diesen Trends entgegen. Der Anteil an spaltbaren Isotopen im Plutonium nimmt bei einer Zunahme des Entladeabbrands ab.

Aus der Sicht der Schonung von Energie Ressourcen ist eine Wiederaufbereitung der abgebrannten Kernbrennstoffe vorteilhaft. Dabei wird Plutonium Rezyklierung im DWR bereits in Frankreich und Deutschland erfolgreich praktiziert. Bei diesen Anwendungen werden 30 bis 50% der BE in einem bestehenden UOX-Kern durch $(UPu)O_2$ (MOX) BE ersetzt. Bei den Untersuchungen im FZK bestand längere Zeit Unklarheit über die Äquivalenzkriterien für UOX- und MOX-BE im gleichen DWR Kern. In der vorliegenden Arbeit wird ein solches Kriterium vorgeschlagen. Es beruht auf Erfahrungen mit dem Einsatz von MOX-BE in französischen und deutschen DWR und wurde auch durch eigene Gesamtkernrechnungen für Voll-MOX Kerne bestätigt. Die Analysen der Kühlmitteldichte Reaktivitätskoeffizienten zeigen, daß der Anteil an spaltbarem Plutonium im MOX $\approx$6% nicht überschreiten soll, wenn die Sicherheitseigenschaften des Reaktors beibehalten werden sollen. Die Plutonium Mehrfach-Rezyklierung wurde für ein Szenarium mit einem Pool gleicher DWR eingehend untersucht. Bei diesem Modell sind am Anfang nur UOX-Kerne vorhanden. Sobald im Pool genügend Plutonium erzeugt worden ist, werden UOX-Kerne durch MOX-Kerne ersetzt. Bei den Rechnungen wird 10 Jahre Umlaufzeit angenommen. Das in MOX-Kernen erzeugte Plutonium wird mit dem Plutonium der noch im Pool vorhandenen UOX-Kerne vermischt. Der Spaltstoff Anteil des Plutoniums im MOX Brennstoff wird auf 6% begrenzt; falls erforderlich wird angereichertes U^{235} verwendet, um die Kritikalitätsanforderungen zu erfüllen. Dieses Szenarium führt nach etwa 60 bis 80 Jahren zu einem quasi-stationären Plutonium Inventar auf einem Niveau von etwa der Hälfte des Falles ohne Rezyklierung zu diesem Zeitpunkt. Der Aufbau von Neptunium beträgt 5..6% des Plutoniums in einem Zyklus und wird praktisch nicht durch die Rezyklierung beeinflußt. Die Americium Menge dagegen erreicht bei der Rezyklierung die gleiche Größenordnung wie Neptunium, bleibt ohne Rezyklierung aber deutlich geringer.

Bei einem geschlossenen Brennstoffkreislauf mit Plutonium Rezyklierung werden große Mengen an rezykliertem Uran (RU) produziert. Vorläufige Untersuchungen zeigen, daß RU auch in DWRs verwendet werden kann. Die erforderliche Anreicherung des RU kann durch spezielle RU-Anreicherungsanlagen oder durch Vermischung des RU mit hoch angereichertem frischen Uran erzielt werden.

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1 Introduction.

During the power generation in light water reactors with uranium fuel the neutron capture in U^{238} principally leads to the production of Np^{239} with a short decay time to Pu^{239}. Neutron capture in secondary heavy isotopes then cause the buildup of further transurania. The back-end of these transurania is one of the key problems of nuclear power generation nowadays. On the one hand the main part of it is fissionable at thermal energies and can be reused in light water reactors. On the other hand the recovery of the transurania from spent fuel needs a more expensive closed fuel cycle. At present direct disposal and reprocessing of spent fuel are two options which may be envisaged with equal priority in Germany (Amendment of the Nuclear Law 1994).

Another important question is the long term radiological impact of these transurania isotopes, having decay half-lifes of many thousands of years. Many investigations are in progress in order to reach a reduction of the radiological consequences of nuclear energy production. Both established systems like light water and fast breeder reactors and more futuristic systems like accelerator driven subcritical blankets are being considered for incineration of transurania and fission product isotopes.

The present study describes investigations related to the reduction of transurania isotopes during nuclear energy production with Pressurized Water Reactors. Transurania isotopes buildup may be reduced by the following means:

1. **Reduction of the conversion ratio from fertile to fissile isotopes in the fuel.** In PWRs this may be realized by the application of better thermalized neutron spectra, e.g. by the use of higher moderator partitions in the reactor lattices (widening of the lattice). Another possibility is the replacement of U^{238} by an alternative resonance absorber material in order to avoid the buildup of Pu^{239} but still keep a sufficiently high Doppler coefficient.

2. **Increasing the burnup of UOX fuel assemblies**, leading to increased plutonium burnup in the uranium core. The consequence of this higher plutonium burnup is an increased production of neptunium, americium and curium isotopes. Moreover, the content of the fissionable isotopes Pu^{239} and Pu^{241} in the unloaded plutonium decreases with increasing fuel burnup.

3. **Plutonium recycling in PWRs.** This option requires a closed nuclear fuel cycle with sufficient recycling capabilities.

In this study burnup increase and plutonium recycling will be investigated. Conversion ratio decrease by lattice modifications or substitution of U^{238} in the fuel will be treated in a separate investigation. The following topics will be considered in more detail.

In chapter 2 the main characteristics of the transurania isotopes will be discussed.

Chapter 3 treats the fundamental effects with the help of basic cell calculations. A short description of the applied calculational models and tools is given. Some parametric studies

for UOX and MOX lattices are performed. The specific buildup per unit of generated energy in PWRs with UOX fuel is investigated.

The problem of equivalency of UOX and MOX fuel assemblies in the same core is investigated in some detail.

In chapter 4 whole core calculations are presented. These calculations are based on reactor lattices of modern PWRs. In a first approximation the fuel assembly cross sections are directly taken from cell calculations. The counterbalancing effects of irregular watergaps and structures in the fuel assembly are neglected.

The full core calculations are performed with the calculational procedures developed for an advanced pressurized water reactor with tight lattices described in reference [1]. Treedimensional diffusion calculations for full MOX cores with hexagonal fuel assemblies are applied.

The applicability of the procedures is confirmed by exploratory calculations for UOX fuel lattices of actual running PWRs. Several scenarios for plutonium multi-recycling in PWRs are described.

In chapter 5 the recycling of selfgenerated plutonium in a pool of identical PWRs is investigated in some detail. As soon as enough plutonium is available, an UOX core is replaced by a full MOX core. After a relatively small number of recyclings the available plutonium in such a system contains a considerable amount of the non-fissile isotopes Pu^{238}, Pu^{240} and Pu^{242}. In the PWR spectrum this requires high fractions of plutonium in the fuel to reach the target fuel burnups, which may lead to undesirable coolant density reactivity coefficients. In the present study this problem is solved by keeping the fissile plutonium fraction Pu_{fis} below $\approx$6% and using enriched uranium if required for criticality reasons.

In chapter 6 the massflows of the transurania neptunium, plutonium, americium and curium are studied in dependence of the target burnups. Both results of parametric cell calculations and of long term investigations for pools of PWRs are presented.

Finally in chapter 7 the possible use of reprocessed uranium (RU) in PWRs is investigated.

2 Characteristics of transurania.

During the irradiation of uranium neutron captures lead to transurania isotopes with very different characteristics. In table 1 a number of properties of these isotopes are summarized. The following aspects are considered.

1. **Lifetime.** The lifetime of the generated isotopes varies from very shortlived (seconds) to very longlived (thousands of years). The characteristic parameter is the half-life, being the time needed to reduce the concentration by decay processes to half its initial value. The tabulated data were taken from reference [2].

2. **Fissionability.** The most effective way to incinerate transurania is fission. Energy production and fission products are very similar for fission of all heavy isotopes. Table 1 contains onegroup fission cross sections, generated by a standard cell calculation for a UOX lattice of a modern PWR with 3.5% U^{235} enrichment.

3. **Incinerability.** Incineration may be realized by other neutron capture processes like $\sigma_c = \sigma_{(n,\gamma)} + \sigma_{(n,\alpha)} + \sigma_{(n,p)}$ and $\sigma_{(n,2n)}$.
   Table 1 contains onegroup cross sections for these processes from the same cell calculations as specified for the fission cross sections.

4. **Source of the heavy isotopes.** The last column of table 1 specifies the main origins of the single heavy isotopes. They have been determined from the calculations mentioned before.

No further analysis concerning hazard potential (e.g. toxicity, mobility) is performed here.

For isotopes without onegroup cross sections in table 1 no data was available for the reactivity calculations with standard 69-group libraries. In these cases the depletion calculations are performed with onegroup data from special depletion libraries, derived from the KORIGEN code system [3], see also reference [1].

The lacking data does not play an important role for usual reactor depletion calculations. Special investigations, e.g. for curium buildup, may need an extension of the standard 69-group libraries with additional isotopes like Cm^{243} and Cm^{245}.

The columns 2 and 3 of table 1 show that most of the listed isotopes undergo α-decay with strongly differing half-lifes.

Short half-lifes, e.g. of Pu^{238}, Cm^{243} and also Am^{241} mean high radiation intensity with consequences for manufacturing. This may lead to the requirement for expensive remote fabrication techniques.

The long decay times cause long term radiological hazard sources with difficult to estimate risks.

Table 1 also shows, that only the isotopes Pu^{239} and Pu^{241} have favourable fission properties. All other isotopes mainly transfer within table 1.

This means, that only a few isotopes have to be considered for long term investigations.

1. For the element **neptunium** the isotope Np^{237} with $T_{\frac{1}{2}}$=2.14 10^6 years.

2. For the element **plutonium** the isotopes Pu^{239} with $T_{\frac{1}{2}}$=2.411 10^4 years, Pu^{240} with $T_{\frac{1}{2}}$=6550 years and Pu^{242} with $T_{\frac{1}{2}}$=3.763 10^5 years. The isotope Pu^{238} causes problems due to its α-radiation and heat generation during fuel fabrication and decays with $T_{\frac{1}{2}}$=87.74 years to U^{234}. The isotope Pu^{241} decays by β^--radiation with $T_{\frac{1}{2}}$=14.4 years to Am^{241}.

3. For the element **americium** the isotope Am^{243} with $T_{\frac{1}{2}}$=7370 years. Am^{241} with $T_{\frac{1}{2}}$=432.6 years needs special caution.

4. For the element **curium** the isotope Cm^{245} with $T_{\frac{1}{2}}$=8500 years. The isotopes Cm^{242} with $T_{\frac{1}{2}}$=162.8 days, Cm^{243} with $T_{\frac{1}{2}}$=28.5 years and Cm^{244} with $T_{\frac{1}{2}}$=18.11 years decay by α-radiation to Pu^{238}, Pu^{239}, Pu^{240}.

On the basis of this analysis mainly all plutonium isotopes and Np^{237}, Am^{241} and Am^{243} are considered in more detail.

Isotope	$T_{\frac{1}{2}}$ ‡	E_α (MeV)	1-gr. cross section (barn)			Most important sources in a PWR
			$<\sigma_f>$	$<\sigma_c>$	$<\sigma_{n,2n}>$	
U^{234}	2.47 10^5 a	4.7	-	-	-	natural isotope, Pu^{238} decay
U^{235}	-	-	47.5	10.7	4.3 10^{-3}	natural isotope
U^{236}	2.342 10^7 a	4.5	0.19	8.5	4.2 10^{-3}	$U^{235}(n,\gamma), Pu^{240}$ decay
U^{237}	6.75 d	-	-	-	-	U^{238} (n,2n), $U^{236}(n,\gamma)$
U^{238}	-	-	0.01	0.91	7.5 10^{-3}	natural isotope
U^{239}	23.5 m	-	-	-	-	$U^{238}(n,\gamma)$
Np^{236}	22.5 h	-	-	-	-	Np^{237} (n,2n)
Np^{237}	2.14 10^6 a	4.8	0.48	35.4	2.5 10^{-3}	Am^{241} and U^{237} decay
Np^{238}	2.117 d	-	-	-	-	Am^{242m} decay, $Np^{237}(n,\gamma)$
Np^{239}	2.355 d	-	0.58	14.2	1.0 10^{-3}	U^{239} and Am^{243} decay
Pu^{236}	2.851 a	5.8	-	-	-	Np^{236} decay, Pu^{238} (n,3n)
Pu^{238}	87.74 a	5.5	2.45	34.9	2.5 10^{-3}	Pu^{239} (n,2n) Np^{238} and Cm^{242} decay
Pu^{239}	2.411 10^4 a	5.2	119.9	67.7	4.3 10^{-3}	Np^{239} decay, $Pu^{238}(n,\gamma)$
Pu^{240}	6550 a	5.2	0.57	228.9	2.0 10^{-3}	$Pu^{239}(n,\gamma)$, Pu^{241} (n,2n) Cm^{244} decay
Pu^{241}	14.4 a	-	122.2	46.8	9.4 10^{-3}	$Pu^{240}(n,\gamma)$, Pu^{242} (n,2n)
Pu^{242}	3.763 10^5 a	4.9	0.40	30.0	3.5 10^{-3}	$Pu^{241}(n,\gamma)$, Am^{242} decay
Pu^{243}	4.956 h	-	-	-	-	$Pu^{242}(n,\gamma)$
Pu^{244}	8.26 10^7 a	4.6	-	-	-	$Pu^{243}(n,\gamma)$
Am^{241}	432.6 a	5.5	1.35	127.8	2.5 10^{-3}	Pu^{241} decay, $Am^{242}(n,2n)$
Am^{242m}	141 a	-	736.5	149.9	2.5 10^{-3}	$Am^{241}(n,\gamma*)$
Am^{242}	16 h	-	736.5	149.9	2.5 10^{-3}	$Am^{241}(n,\gamma)$, Am^{243} (n,2n)
Am^{243}	7370 a	5.3	0.42	51.0	2.5 10^{-3}	$Am^{242}(n,\gamma)$, Pu^{243} decay
Cm^{242}	162.8 d	6.1	1.19	4.4	0.2 10^{-3}	Am^{242} decay, $Cm^{243}(n,2n)$
Cm^{243}	28.5 a	5.8	-	-	-	$Cm^{242}(n,\gamma)$, Cm^{244} (n,2n)
Cm^{244}	18.11 a	5.8	0.96	15.4	2.5 10^{-3}	$Cm^{243}(n,\gamma)$
Cm^{245}	8500 a	5.4	-	-	-	$Cm^{244}(n,\gamma)$
Cm^{246}	4730 a	5.4	-	-	-	$Cm^{245}(n,\gamma)$
Cm^{247}	1.56 10^7 a	4.9	-	-	-	$Cm^{246}(n,\gamma)$

‡ a years, d days, h hours, m minutes

Table 1: Characteristics of transurania isotopes.

3 Basic investigations.

In this section some principal investigations on the basis of lattice cell calculations will be presented. The applied calculational procedures have been developed for the analysis of advanced pressurized water reactors (APWR) with tight lattices, see reference [1]. Parametric calculations for UOX lattices and some comparisons with actual MOX lattices are discussed.

3.1 Calculational model

Figure 1 shows the applied three-zone Wigner-Seitz lattice cell for the basic calculations. This cell was proposed for a common benchmark on plutonium recycling in PWRs organized by Electricite de France (EDF) and Forschungszentrum Karlsruhe (FZK) [4]. It is derived from a standard French 900MWe PWR-design and is representative for modern PWRs.

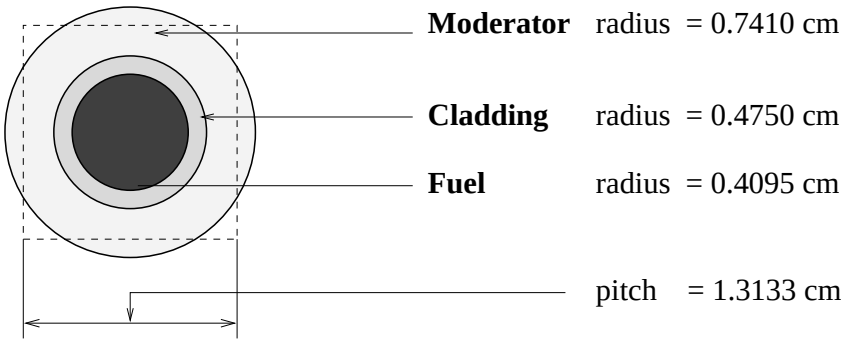

Figure 1: Cell geometry for PWR lattice calculations, T=20^oC

The main characteristics are

- Can material zircalloy,
- Rod diameter 9.5 mm, canning thickness 0.0655 mm,
- Fuel weight $\approx$4.75 g/cm,
- Square lattice with p/d$\approx$1.38,
- Coolant density 0.71 g/cm^3.

These lattice specifications will be used in all subsequent cell calculations.

3.2 Applied calculational procedures.

The applied calculational procedures have been developed for the analysis and the design of advanced pressurized water reactors with tight lattices. In reference [1] more detailed information may be found. The main features are:

- The calculational procedures of the so-called **KA**rlsruhe **R**eactor **BU**rnup **S**ystem KARBUS [1] are integrated in the modular **KA**rlsrhe **PRO**gram **S**ystem KAPROS [6], primarily designed at FZK for fast reactor investigations. For the present work recent KAPROS versions have been used. Based on the standard KAPROS3-kernel [7] for the IBM-MVS operating system a KSSKUX-kernel for UNIX workstations has been developed. Starting from this KSSKUX-kernel a new system for use with the IBM-MVS-XA extended architecture option has been created: the KSSKXA-kernel. The latter enables the use of core storage larger than about 10MB as limitation in the older system.

- The KARBUS procedures combine advantages of existing established special purpose procedures for thermal and fast reactors.

- For the lattice calculations first collision probability methods are implemented. These are based on the formalisms of the well-known English WIMS code [8].

- The group constant libraries use the flexible GRUBA format [9], developed at FZK. For the treatment of temperature dependent neutron upscattering special extensions to the original fast reactor oriented options have been implemented. Contrary to the WIMS code, the GRUBA library options enable the treatment of neutron transfer between energy groups with the storage of distinct transfer matrices for every important neutron reaction, e.g. elastic and inelastic scattering, $(n,2n)$ and $(n,3n)$ reactions. The original WIMS code only enables the storage of one transfer matrix for each material.

- Additional to the fast reactor libraries with energy group structures with 26 and 208 groups, libraries with 69 and 334 energy group structures with better energy resolution in the lower energy region below 4 eV have been established. For the energy resolution below 4 eV the group structure of the WIMS code was adopted.

- Several improvements in the energy region with neutron resonances have been introduced [1]:

  - The application of the f-factor formalism for the treatment of the selfshielding effect in homogeneous media has been refined with a number of new options.
  - The selfshielding effects in heterogeneous media may be treated with the help of equivalency considerations for homogeneous media.
  - The so-called Bell-factor treatment has been improved by an originally derived energy dependent formula, see the references [1] and [10].

  - Alternatively to the fast f-factor formalisms more accurate and more computer time consuming fine-flux methods have been made available, see reference [1].

- Extensive validation investigations have been performed to qualify the application of KARBUS for the description of reactors with thermal, epithermal and fast neutron spectra. This validation work was based both on experimental data and on theoretical benchmark exercises.

3.3 Parametric investigations for UOX lattices.

In order to obtain a consistent database for further evaluations, a number of parametric investigations for UOX lattices have been performed. Standard cell lattice calculations, using the 69-group library G69P1V03, see reference [1], have been applied. Additionally to the validation work in reference [1] these calculational procedures were recently compared with the standard calculational procedures at EDF in a common benchmark investigation of EDF and FZK for plutonium multi-recycling in PWRs [4, 5]. The results of these comparisons are very satisfactory. For the lattice of figure 1 the enrichment of U^{235} in the UOX fuel has been varied from 3.2% to 4.5%. Burnup calculations have been carried out for burnups up to $\approx$70 gigawatt days per ton of heavy metal (GWD/THM). For the burnup calculations as a function of the fuel burnup, cycle time and power rating are dependent parameters. Most calculations were performed with a value of 164 watt per cm (W/cm). The influence of the power rating on transurania burnup was investigated by a parameter variation from 120 to 200 W/cm for the case of 4% U^{235} enrichment. The main results of this analysis are shown in figure 2. K_∞ and the buildup of neptunium, plutonium and americium are plotted as a function of burnup. We may observe, that the influence of the power rating is small in most cases. Only the Am^{241} buildup shows a somewhat larger sensitivity.

The figures 3 to 5 show the main results of the parametric calculations. In figure 3 K_∞ and the buildup of plutonium, Np^{237} and Am^{243} are plotted as a function of the fuel burnup. As to be expected, K_∞ depends strongly on the U^{235} enrichment of the fuel. The plutonium buildup is not very sensitive to this parameter. The results for Np^{237} and Am^{243} show a decrease of the buildup for increasing U^{235} enrichment. This can be explained by an increase of the partition of the plutonium fission to the total fissions in the fuel for cases with smaller U^{235} enrichment for the same target burnup of the fuel. The figures 4 and 5 show the end of life (EOL) composition of the plutonium for different U^{235} enrichments and for different fuel burnups. The decrease of the fissile isotope Pu^{239} may be observed clearly. The non-fissile isotopes Pu^{238} and Pu^{242} show a steady increase. For the non-fissile isotope Pu^{240} and the fissile isotope Pu^{241} a saturation behaviour may be observed, caused by balancing production and destruction effects.

3.3.1 Influence of the fuel cycle ex-core times.

In a closed fuel cycle finite ex-core times are inevitable. Between the reactor shutdown and the reuse of the recycled plutonium in a next reactor core two stages may be distinguished:

1. **Cooling time.** Time between the end of irradiation and the chemical separation of the plutonium during the fuel reprocessing. After reactor shutdown several years of time delay are required to reduce the heat and radiation level of the spent fuel assemblies by the decay of the short-lived fission products. The main fuel changes during this period are the buildup of Pu^{238} from the α-decay of Cm^{242} and of Pu^{239} from the β-decay of Np^{239}. Further the α-decay of Pu^{238} and the β-decay of Pu^{241} have to be considered.

2. **Fabrication time.** Time between the separation of the plutonium and the startup in the reactor. During this time the decay of Pu^{238} into U^{234} and of Pu^{241} into Am^{241} have to be taken into account. Especially the buildup of Am^{241} in the MOX fuel has an unfavourable influence on its reactivity. For that reason the time between reprocessing and reactor startup should be kept as short as possible.

For a specified cooling time t_1 and fabrication time t_2 the changes in the plutonium fuel composition may be determined with sufficient accuracy by the following formulas:

$$N_{Pu^{238}} = N^0_{Pu^{238}} \cdot e^{\frac{-(t_1+t_2)ln(2)}{T_1}} + N^0_{Cm^{242}} \cdot (1 - e^{\frac{-t_1 ln(2)}{T_3}}) \cdot e^{\frac{-t_2 ln(2)}{T_1}} \tag{1}$$

$$N_{Pu^{239}} = N^0_{Pu^{239}} + N^0_{Np^{239}} \tag{2}$$

$$N_{Pu^{240}} = N^0_{Pu^{240}} \tag{3}$$

$$N_{Pu^{241}} = N^0_{Pu^{241}} \cdot e^{\frac{-(t_1+t_2)ln(2)}{T_2}} \tag{4}$$

$$N_{Pu^{242}} = N^0_{Pu^{242}} \tag{5}$$

$$N_{Am^{241}} = N^0_{Pu^{241}} \cdot e^{\frac{-t_1 ln(2)}{T_2}} \cdot (1 - e^{\frac{-t_2 ln(2)}{T_2}}) \tag{6}$$

$$N_{Np^{239}} = 0. \tag{7}$$

$$N_{Cm^{242}} = 0. \tag{8}$$

with

$$T_1 = T^{Pu^{238}}_{\frac{1}{2}} \; , \; T_2 = T^{Pu^{241}}_{\frac{1}{2}} \; , \; T_3 = T^{Cm^{243}}_{\frac{1}{2}}$$

The validity of these formulas could be verified by comparisons with more accurate KARBUS calculations.

In the references [4, 5] a cooling time of 3 years and a fabrication time of 2 years were chosen for the EDF/FZK benchmark investigations on plutonium recycling in PWRs. These are expected values for a settled closed fuel cycle. For the present study less optimistic delay times are used. For a longer transition phase values of 7 years cooling and 3 years fabrication time are expected to be more reasonable. Figure 6 shows the plutonium fuel compositions after 7 years cooling and 3 years fabrication time as a function of U^{235} enrichment of the UOX fuel and of the fuel burnup. A direct comparison with the end of life fuel in figure 4 shows a significant decrease of the Pu^{241} fraction, caused by its rather short half-life of 14 years, and an additional small fraction of Am^{241}, also caused by the Pu^{241} decay since the chemical separation of the plutonium during the fuel reprocessing.

3.3.2 Transurania buildup at constant energy production.

In this section some estimates for the buildup of transurania per unit of energy production with PWRs with UOX fuel and different discharge burnups are performed. For a constant thermal power P_{th} the fuel inventory G is determined by the linear power rating P_L and the specific fuel weight per cm fuel rod ρ_L.

$$G = \frac{P_{th} \cdot \rho_L}{P_L} \tag{9}$$

The fuel burnup BU for T equivalent full power days (EFPD) and N burnup cycles amounts to:

$$BU = \frac{P_{th} \cdot T \cdot N}{G} \tag{10}$$

From (9) and (10) follows:

$$BU = \frac{P_L \cdot T \cdot N}{\rho_L} \tag{11}$$

A further crucial boundary condition is the reactor criticality. The following condition is required at the end of a reactor cycle after T EFPD:

$$k_{eff,EOC}^{Reactor} = 1. \tag{12}$$

The choice of the linear power rating and of the burnup cycle time mainly determine the needed excess reactivity at reactor startup and in this way the required fuel enrichment.

The production of a certain amount of energy per unit time with different discharge burnup may be realized in several ways:

1. **Changing the number of burnup stages in a core.** For a given enrichment with a corresponding burnup cycle time, the fuel discharge burnup will increase if the number of fuel batches in the core increases.

2. **Changing the enrichment of the fuel.** At a given power rating level the cycle time and mean cycle burnup will increase if the fuel enrichment is increased.

The number of burnup stages N in a core determines the ratio between the mean discharge burnup Bu_{EOL} at end of life and the cycle burnup Bu_{EOC} in one burnup cycle. In a first approximation the following formula may be applied for the ratio between Bu_{EOC} and Bu_{EOL}:

$$\frac{Bu_{EOC}}{Bu_{EOL}} = \frac{N+1}{2N} \tag{13}$$

Figure 7 shows a graphical representation of equation (13). We may observe, that the burnup gain decreases rapidly with increasing number of burnup stages.

The tables 2 to 4 summarize results for transurania buildup at the end of the corresponding reactor cycles (EOC). The decay of Np^{239} to Pu^{239} with a half-life of 2.4 days should be kept in mind. In practical cases Np^{239} can be added to the plutonium, see equation 2.

Table 2 gives the buildup of neptunium, plutonium and americium for 4 U^{235} enrichments (3.2, 3.5, 4 and 4.5%) and 2 discharge burnups (33 and 50 GWD/THM). For a three batch fuel management 3.2% enrichment is in accordance with experiences with modern PWRs to reach 33 GWD/THM discharge burnup. The planned extension of the discharge burnup to 50 GWD/THM is expected to be realizable with a three batch fuel management and ≈4.5% enrichment [4]. The last row of table 2 shows the amount of remaining U^{235} in the spent fuel. For all cases this value is slightly below 1%.

Table 3 gives the plutonium compositions for the same parameter as in table 2.

In table 4 some estimates for the normalized transurania buildup in PWRs are presented. For the normalization the results for the transurania buildup at 33 GWD/THM discharge burnup are multiplied by a factor of 1.5 to obtain the same energy production per time unit as for the discharge burnup 50 GWD/THM. The columns 2 to 4 compare buildup for the same enrichment of 3.5% U^{235}. The discharge burnup of 50 GWD/THM with 3.5% U^{235} is not realistic for full burnup charges in a PWR. The comparison is intended to show the possible influences of the U^{235} enrichment on the buildup of neptunium and americium. The columns 5 to 7 compare buildup for different enrichments of 3.2% U^{235} at 33 GWD/THM and 4.5% U^{235} at 50 GWD/THM. As mentioned before these cases are realistic for a three batch fuel management in modern PWRs. We can observe, that the specific production of Np^{237}, Am^{241} and Am^{243} significantly increases if the discharge burnup is enhanced from 33 to 50 GWD/THM. As a consequence more plutonium with better fissile properties is produced for the lower discharge burnup of 33 GWD/THM. The total amount of transurania decreases with increasing discharge burnup; the transurania produced are partly destroyed at higher burnups with a shift to americium buildup.

3.4 The calculation of MOX lattices.

PWR lattices with MOX fuel may be calculated with the same procedures as in the case of UOX fuel. However, some pronounced resonances in the plutonium cross sections need a careful treatment of resonance absorption calculations in PWR lattices with MOX fuel with high plutonium fractions. In the early stages of calculations for light water moderated lattices with MOX fuel, considerable difficulties have been encountered with the treatment of the large broad resonance of Pu^{240} at 1 eV and with the large resonance of Pu^{242} at 1.67 eV, see for example reference [1] for more details. An important difference between UOX and MOX fuel in PWR lattices shows the reactivity loss per unit burnup. The higher plutonium content leads to harder neutron spectra and to higher conversion ratios from fertile to fissile isotopes, resulting in a less steep slope for the reactivity as a function of burnup. Together with the different cross sections of the main fissile materials this raises the problem of equivalency of UOX and MOX fuel assemblies in the same PWR core.

3.5 Equivalency of UOX and MOX lattices in a PWR core.

At the level of cell calculation no simple clear prescription exists for the definition of equivalency of UOX and MOX fuel assemblies in a PWR core. In fact only the whole core behaviour can give decisive answers. Reactivity and thermodynamic related boundary conditions must be fulfilled during the whole planned reactor cycle.

Early investigations used the arbitrary criterion to apply the **same** value of K_∞ at the end of cycle condition in UOX and MOX lattices [11]. For discharge burnups of 50 GWD/THM this procedure produced higher values for the required Pu_{fis} content of the MOX fuel, compared to the benchmark results of reference [4]. A more detailed analysis of this problem showed that the criterion mentioned before seems to be too conservative for the determination of a realistic Pu_{fis} content of MOX fuel assemblies to be loaded in UOX PWR cores:

1. The first indication comes from the history of the EDF/FZK benchmark on plutonium recycling in PWRs. Indeed in the first specification the above mentioned criterion was proposed (equal K_∞ at EOC). The final benchmark specifies a value of K_∞ at EOC that is based on experiences with MOX fuel assemblies with discharge burnups of ≈33 GWD/THM in French 900 MWe PWRs. For the benchmark discharge burnup of 50 GWD/THM the plutonium fraction in the MOX fuel must be chosen in a such a way, that the K_∞ value is the same as for the MOX fuel applied in the 900 MWe French PWRs. The difference at the end of cycle condition for the K_∞ values for the applied UOX and MOX fuel amounts to

$$\Delta K_\infty = K_\infty^{UOX} - K_\infty^{MOX} \approx 0.03 \tag{14}$$

2. Investigations at FZK with the KARBUS code for the KKP2 1300 MWe PWR at Philippsburg showed a similar behaviour as expressed in formula (14). Figure 8

presents the K_∞ curves as a function of burnup for the UOX and MOX lattices as been used in KKP2. The less steep slope of the reactivity curve as a function of the burnup for MOX lattices compared to UOX lattices clearly may be observed. The difference in K_∞ at 22 GWD/THM, being $\frac{2}{3}$ of the end of cycle burnup 33 GWD/THM from formula (13) for 3 batches and approximately the average core burnup before reloading, amounts to $\approx$0.03.

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	9.592	9.581	9.561	9.537	11.733	11.777	11.855	11.932
Np^{237}	0.4254	0.4306	0.4368	0.4408	0.6453	0.6650	0.6916	0.7119
Np^{239}	0.0883	0.0846	0.0791	0.0745	0.1002	0.0965	0.0907	0.0854
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
U^{235}	95.596	95.596	95.597	95.598	93.618	93.613	93.604	93.595

Table 2: Transurania buildup, power rating 164 W/cm, EOC data

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	1.50	56.27	22.35	14.15	5.72	70.42	2.89	47.25	24.72	14.85	10.30	62.10
3.5	1.46	57.79	21.61	14.00	5.14	71.79	2.86	48.44	24.23	14.96	9.51	63.40
4.0	1.38	60.15	20.45	13.67	4.34	73.82	2.79	50.45	23.37	15.06	8.33	65.51
4.5	1.31	62.31	19.39	13.29	3.69	75.60	2.71	52.43	22.49	15.07	7.30	67.50

Table 3: Plutonium compositions, power rating 164 W/cm, EOC data

Material (kg/TIHM)	U^{235} (%)		Diff. (%)	U^{235} (%)		Diff. (%)
	3.5	3.5		3.2	4.5	
	Burnup (GWD/THM)			Burnup (GWD/THM)		
	33 (x1.5)	50		33 (x1.5)	50	
Pu	14.372	11.777	+22.0	14.389	11.932	+20.6
Np^{237}	0.6459	0.6650	-2.9	0.6381	0.7119	-10.4
Np^{239}	0.1268	0.0965	+31.4	0.1325	0.0854	+55.1
Am^{241}	0.0559	0.0565	-1.0	0.0555	0.0638	-13.0
Am^{243}	0.1304	0.2914	-55.3	0.1503	0.2095	-28.3
Transurania	15.331	12.887	+15.9	15.366	13.003	+15.4

Table 4: Transurania buildup normalized to the same net energy production, EOC data

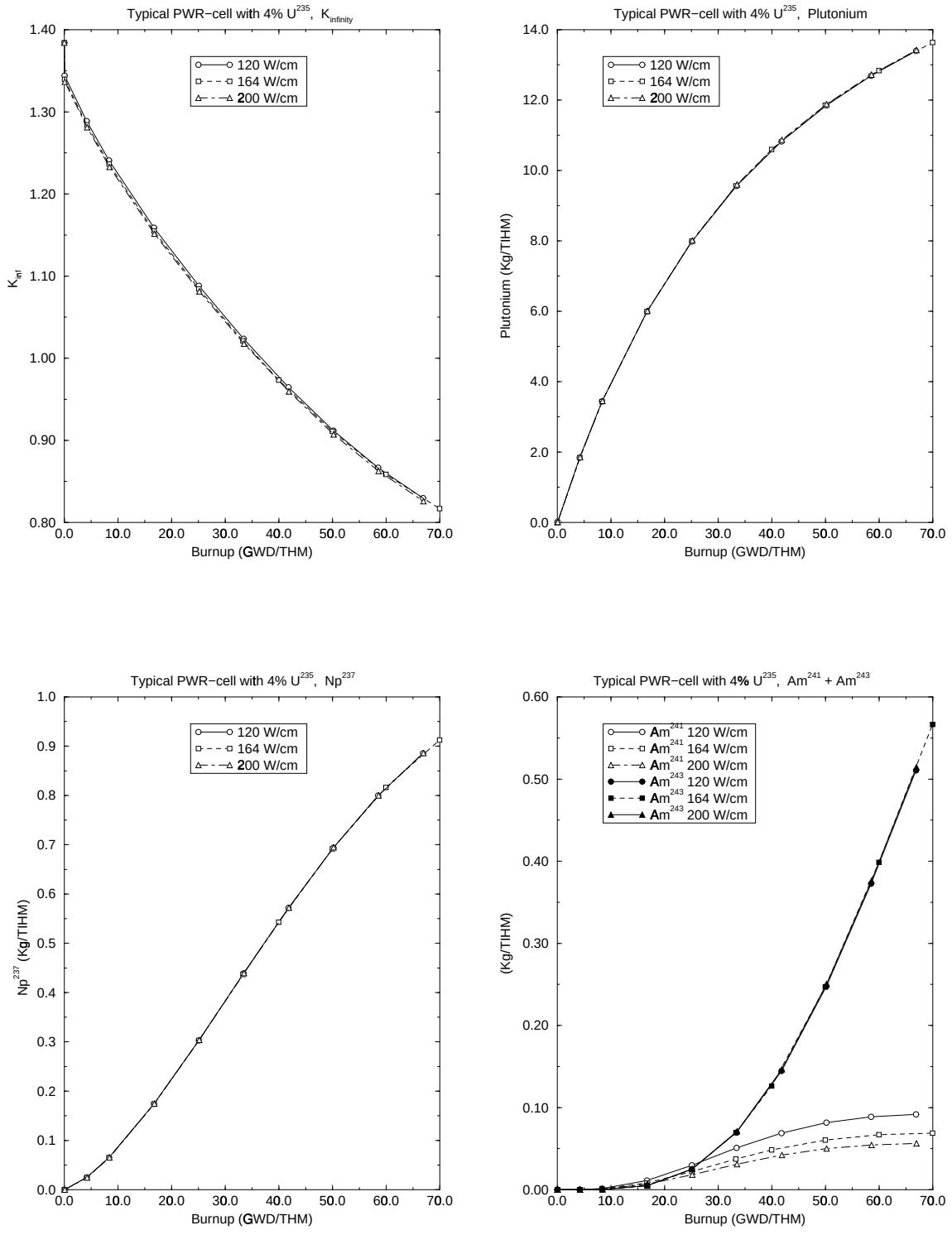

Figure 2: Influence of power ratings on the buildup of transurania.

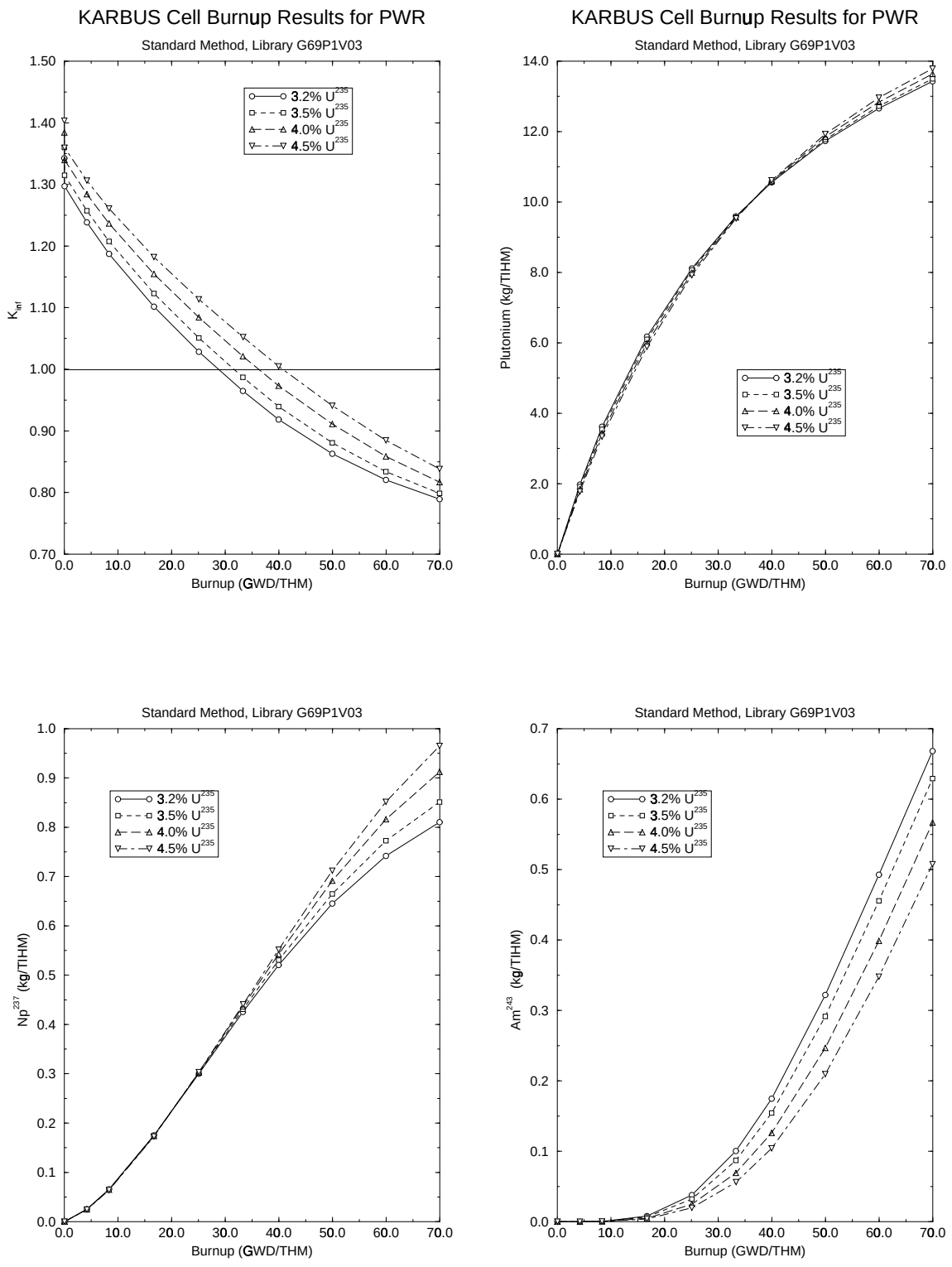

Figure 3: Influence of the U^{235} enrichment on the buildup of transurania.

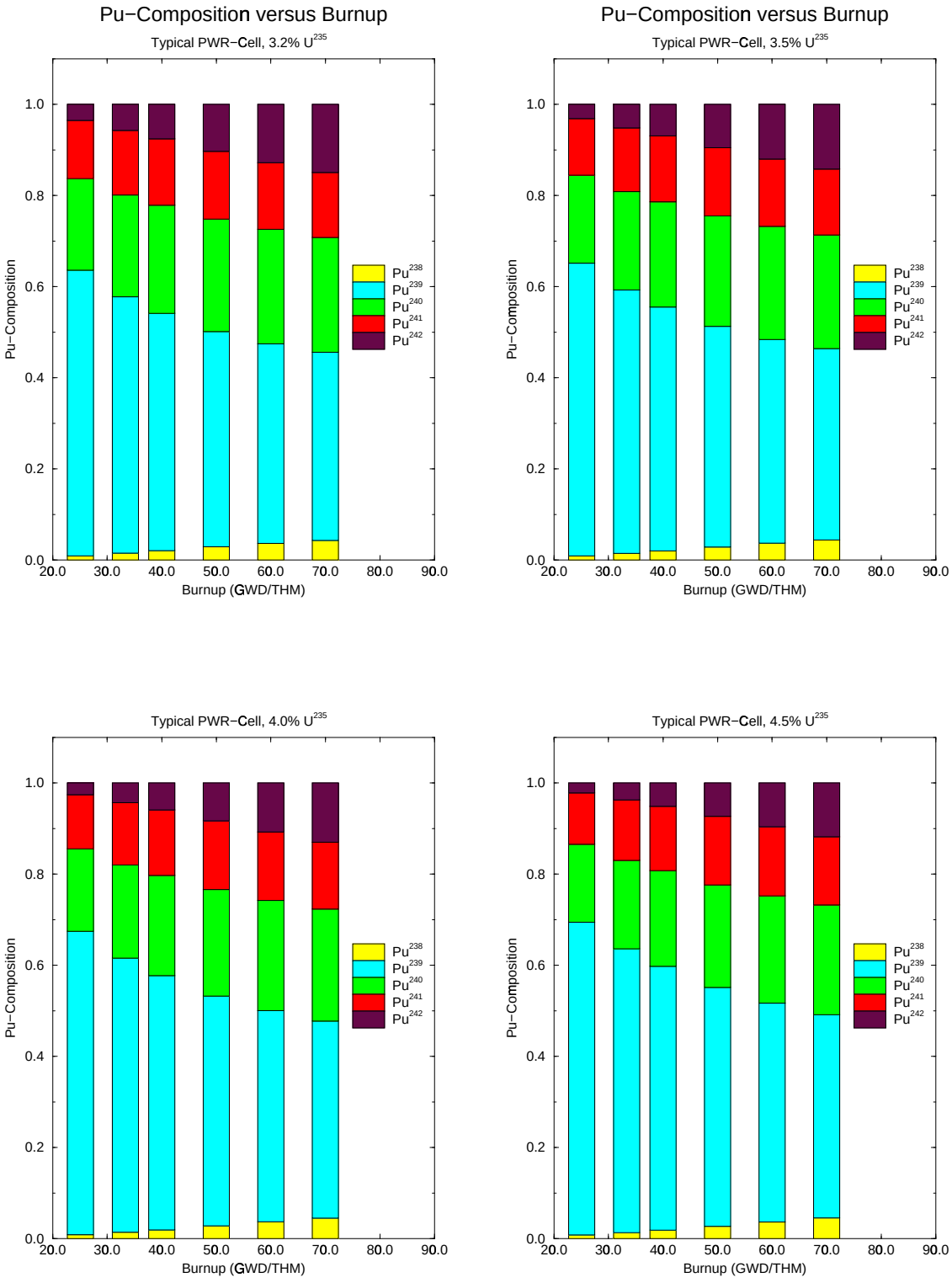

Figure 4: Influence of the U^{235} enrichment on the EOL plutonium composition.

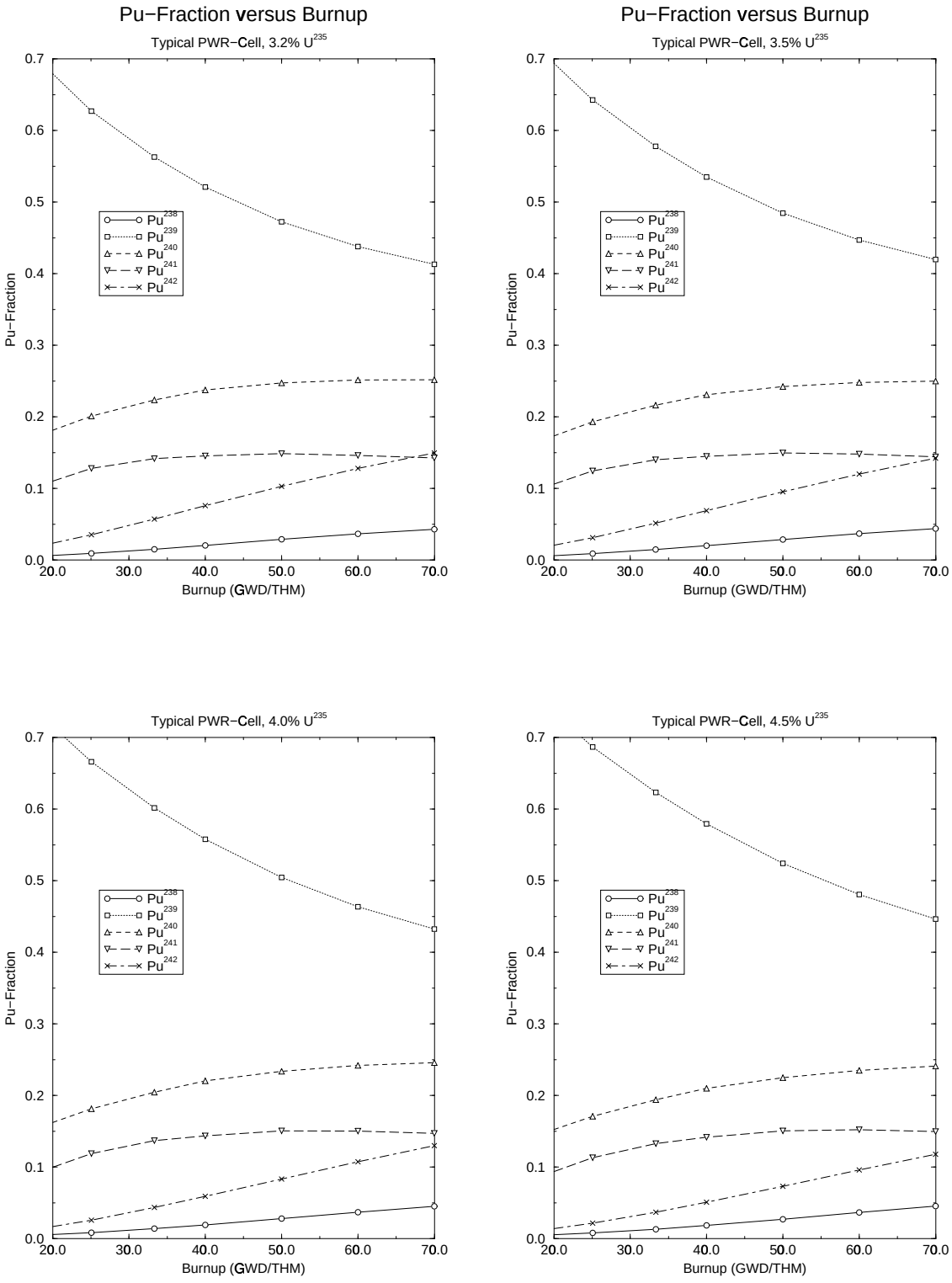

Figure 5: Influence of the U^{235} enrichment on the EOL plutonium composition.

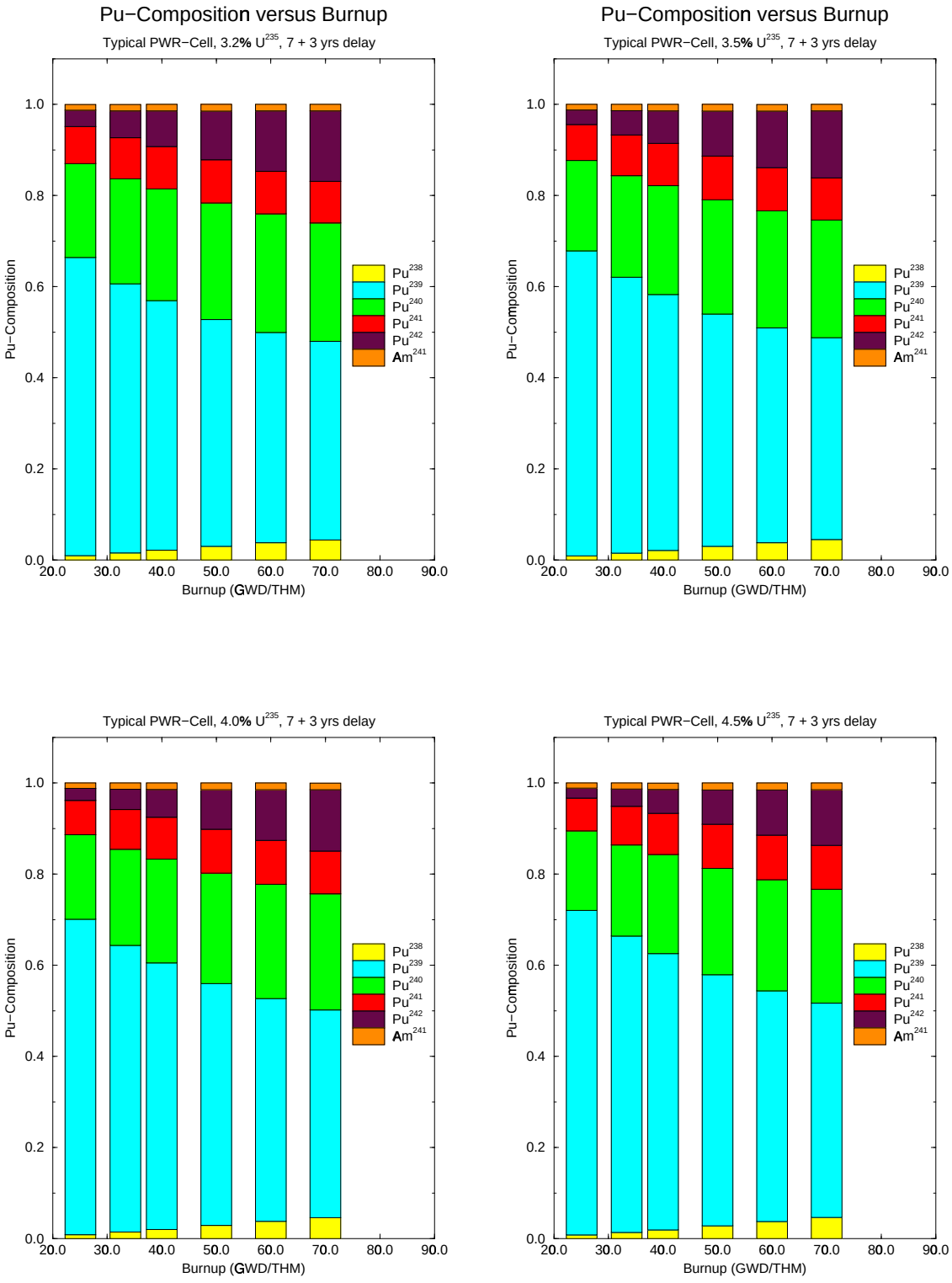

Figure 6: Influence of the U^{235} enrichment on the plutonium composition after 7 years cooling and 3 years fabrication time.

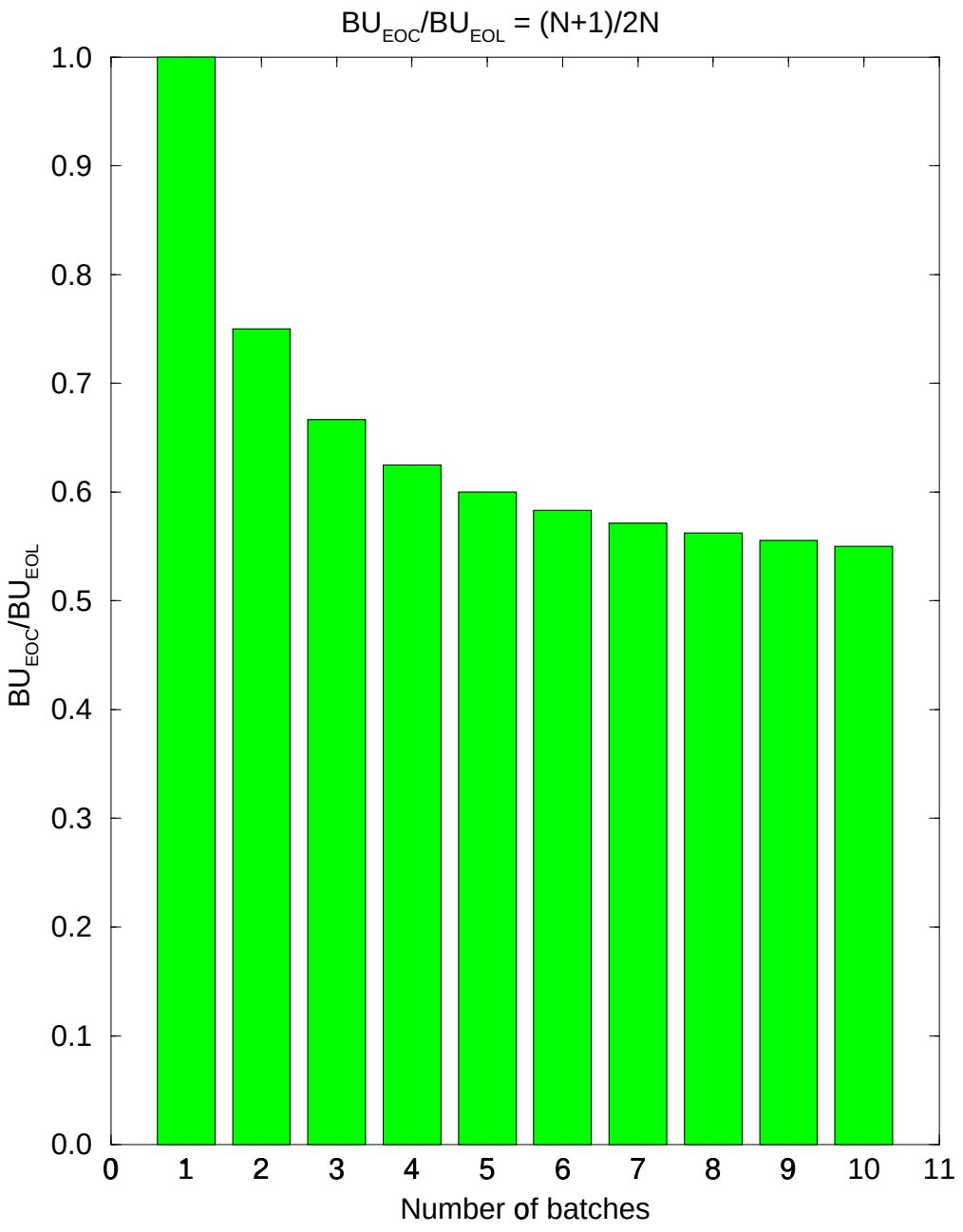

Figure 7: Ratio burnup at EOC to EOL vs the number of core burnup stages.

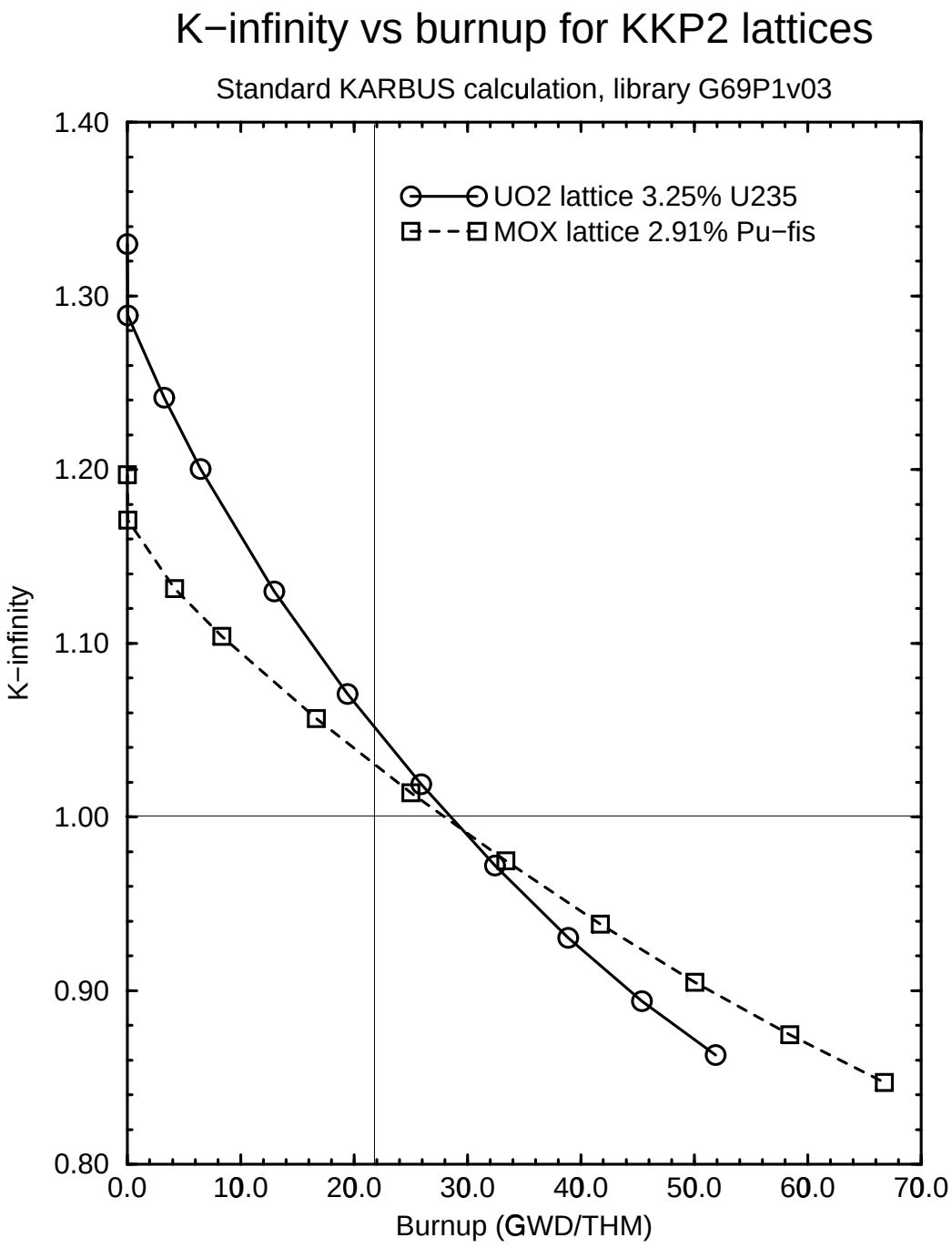

Figure 8: Comparison of K_∞ values for UOX und MOX fuel lattices in KKP2

4 Plutonium recycling and whole core investigations.

In order to obtain reliable estimates for the required plutonium content in the MOX fuel of PWRs, whole core investigations are needed. Such whole core calculations, including fuel management for a number of reactor cycles, are very complex and need large efforts if applied to an operating reactor. At present the use of MOX fuel assemblies in PWRs is state of the art in several countries, e.g. in France and Germany [12, 13]. In selected PWRs up to 33% of the fuel assemblies have been replaced by MOX loadings until now. The replacement of 50% MOX assemblies already has been licensed in Germany. The actual plutonium recycling has been realized with pretty good plutonium with high fissile fractions. This plutonium comes mainly from PWR fuel with discharge burnups slightly above 30 GWD/THM. The fissile fraction is about 70%. The fissile plutonium fraction Pu_{fis} in the MOX fuel assemblies varies in the range of 3 to 4% for target burnups around 33 GWD/THM. Several contributions in reference [14] give more detailed information about MOX exploitation in PWRs.

For the present study not too detailed calculations could be performed. The calculational procedures developed for the investigation of advanced pressurized water reactors with tight lattices, described in reference [1], enable more global investigations, based on equilibrium cores of full MOX PWRs with hexagonal fuel assemblies. These methods have been used for a number of whole core studies for PWRs. The next sections describe the applied whole core calculations.

4.1 Calculational models.

Whole core calculations for PWRs usually are subdivided into a number of subsequent steps. The basic geometry of the fuel in a PWR consists of a regular fuel rod lattice, usually in a square fuel assembly arrangement. The PWR core contains a 193 of such fuel assemblies. In the equilibrium state of a PWR core the composition of the fuel assemblies strongly depends on their irradiation history. Satisfactory power distributions and discharge burnups only may be achieved by careful fuel management procedures, e.g. choice of the number of burnup stages (batches) in the core and of the fuel assembly shuffling strategies. The applied methods from reference [1] have been developed for PWR cores with some futuristic characteristics:

1. Use of **hexagonal fuel assemblies** instead of square ones. As a consequence the fuel lattices have a triangular arrangement.
2. Use of **full MOX** cores.

The main features of the calculational procedures are discussed below.

4.1.1 Lattice calculations.

The lattice calculations are performed with one-dimensional transport calculations in a cylinderized Wigner-Seitz cell. Figure 1 of section 3.1 shows the principle for such a model for

a square reactor lattice. The cell radius is determined from the cell pitch (distance of the fuel rods) by the requirement of equal areas of the cell cross section. For triangular reactor lattices the same methodology is applied. The transport calculations usually are performed with collision probability methods, based on the formalisms of the WIMS code [8]. Alternative control calculations are possible with the one-dimensional discrete ordinate transport code ONETRA [15], a KAPROS version of the program ONETRAN [16]. The difference between square and triangular lattices is taken into account for the determination of the effective resonance cross sections in the fuel zone of the Wigner-Seitz cell. During these calculations the so-called Dancoff factor defines the probability that a neutron born in the single fuel cell will have its next collision in a neighbour cell. The KARBUS calculations utilize geometry dependent Dancoff factors. More detailed informations about the applied lattice calculation methods may be found in reference [1].

4.1.2 Fuel assembly calculations.

Starting from the results of the lattice calculations, the next step of the reactor calculations is the determination of the mean fuel assembly cross sections. The fuel assembly not only consists of the regular reactor lattice, but also of additional material zones like fuel rod spacers, fuel assembly boxes and additional water gaps. In reference [1] several types of fuel assembly models are investigated. The additional materials in the fuel assembly have counterbalancing effects on the overall behaviour. The irregular water gaps lead to spectrum softening with reactivity increase, whereas the additional structural materials give an increase of the parasitic absorptions, leading to a decrease of the fuel assembly reactivity. Detailed analyses have shown, that the basic lattice calculations give a good approximation for the mean fuel assembly cross sections. Therefore for the following exploratory whole core investigations no detailed fuel assembly calculations have been carried out. The mean lattice cross sections have been directly used for the core calculations.

4.1.3 Core calculations.

The whole core calculations of reference [1] are based on hexagonal fuel assemblies with triangular reactor lattices. The main reason for this choice were the tight lattices to be used. In reference [17] the differences between square and hexagonal fuel assemblies are analyzed in some detail. Especially for tight lattices the hexagonal layout has some advantages from the thermodynamical and neutron physics point of view. Moreover hexagonal fuel assemblies in PWRs have proven to be feasible in the Russian VVER reactors [18]. For the exploratory whole core investigations it may be expected, that the fuel assembly layout does not play a significant role. Therefore the available calculation procedures with hexagonal fuel assemblies have been used for the subsequent investigations.

4.1.3.1 Heterogeneous versus homogeneous MOX cores. Until now plutonium recycling in PWRs only has been realized by mixing UOX and MOX fuel assemblies in existing

PWR cores and no definite plans for full MOX cores exist in Germany at present. However, several proposals for future full MOX PWR cores have been made, see for example some contributions in reference [14].

In principle the methods of reference [1] enable the calculation of cores with a mixing of UOX and MOX fuel assemblies.

4.1.3.2 Equivalency between UOX and MOX fuel assemblies. As pointed out in section 3.5 no simple prescriptions exist for the determination of the enrichments of mixed UOX and MOX fuel assemblies in a PWR core. So it would be quite laborious to define a satisfactory fuel management scheme for such theoretical cores.

In order to avoid further complications for the fuel management and because full MOX cores are envisaged for the future, all subsequent whole core calculations will be performed for full MOX cores. It may be expected, that such calculations give reliable information for the needed plutonium enrichments in PWRs.

4.1.4 Fuel management.

For the fuel management in PWRs a number of relevant aspects play a role:

1. **Length of the reactor cycle in equivalent full power days.** Common praxis for PWRs are reloading periods of 1 to 1.5 years with load factors close to 90%.

2. **Number of fuel batches,** i.e. the number of different residence times of the fuel assemblies in the core at a certain time. Values of 3 to 6 batches may be found in PWR investigations. The consequences are discussed in some detail in section 3.3.2. See formula (13) and figure 7.

3. **Fuel assembly shuffling.** At every fuel assembly reloading, a rearrangement of the fuel assemblies in the core may be necessary or useful to obtain satisfactory overall nuclear and thermodynamic core behaviour. Especially the radial form factor of the power distribution must be kept acceptable during the whole reactor cycle. In the existing nuclear power plants large efforts are spent to optimize the fuel management with respect to overall economics.

The present investigations take care of these aspects.

A further significant simplification is introduced by the limitation to study only equilibrium cores. The results of these calculations might be used for the design of transition to equilibrium cores.

4.1.4.1 Characteristics of the equilibrium core. The subsequent whole core investigations are based on equilibrium cores. After a completed equilibrium reactor cycle a constant number of fresh identical fuel assemblies replace such ones which have reached the foreseen number of residence cycles. The now available fuel assemblies are placed into the core with always the same predefined shuffling scheme. For the tight lattice PWR investigations in reference [1] the **A**dvanced **R**eactor **CO**re **SI**mulator code ARCOSI was developed for the iterative determination of such equilibrium cores. In the mean time ARCOSI could be successfully tested for fast reactors and for conventional PWRs. The threedimensional whole core calculations are performed with the nodal code HEXNOD [19], usually with diffusion approximation. ARCOSI enables an arbitrary number of succeeding reactor cycle simulations. Each reactor cycle may be subdivided into an arbitrary number of micro timesteps. At every micro timestep a criticality search by adjustment of the coolant boron concentration is done.

4.1.4.2 The fuel assembly shuffling scheme. For the fuel assembly shuffling the code ARCOSI has a couple of options. Both direct and indirect addressing within the reactor core model may be applied for the shuffling of the assemblies. For the subsequent equilibrium core simulations the method of indirect addressing is used: for every fuel assembly position in the core model, the order of the burnup stage within the available fuel assemblies is fixed.

4.1.5 Sequence for a whole core calculation.

The most important steps during a PWR whole core calculation in this study are:

1. **Choice of the fuel to be utilized.**

2. **Choice of the required discharge burnup.**

3. **Determination of the fissile enrichment of the fuel.** For this task a similar procedure as proposed for the EDF/FZK benchmark on plutonium recycling could be applied. With the help of trial and error estimates for the fissile fuel content in burnup calculations, a predefined K_{∞}^{EOC} value at the end of the reactor cycle must be found. The applied values are for the UOX lattices $K_{\infty}^{EOC} \approx 1.06$ and for the MOX lattices $K_{\infty}^{EOC} \approx 1.03$. Only relatively crude determinations have been performed for the present exploratory investigations. All important results from these calculations are automatically stored on standard KAPROS archives for use in succeeding steps.

4. **Creation of a special purpose library HXSLIB** for use with the ARCOSI code. Usually a HXSLIB library contains 4 group macroscopic cross sections of the fuel assemblies to be used in the whole core calculations, including axial and radial reflector zones. These macroscopic cross sections depend on burnup stage, fuel temperature, coolant density and B^{10} content of the coolant. They are calculated with KARBUS restart options from the KAPROS archives mentioned before. Figure 9 shows the

K_∞ curves of a HXSLIB library for first generation MOX fuel assemblies. The plutonium comes from PWR UOX fuel assemblies with 4.5% U^{235} enrichment and 50 GWD/THM discharge burnup. The Pu_{fis} enrichment in the MOX fuel amounts to 5.5%.

5. **Choice of the number of fuel batches and of the fuel management scheme** for the core simulations with the code ARCOSI. This task may be very laborious. Until now no tools have been developed to automatize this optimisation. The trial and error procedure works satisfactorily with the help of fast graphical evalutions of the most important results. Most of the full core calculations are based on 30^o symmetry in the core. In the axial direction symmetry to the midplane is assumed with 16 axial zones. For the applied core design from reference [1] with 349 hexagonal fuel assemblies, 34 positions describe the radial core layout in the 30^o model. Figure 10 shows an example of the applied 30^o cross section of the calculational model for core burnup simulations. In this case the MOX fuel comes from PWR UOX fuel assemblies with 3.2% U^{235} enrichment and 33 GWD/THM discharge burnup. The target burnup of the MOX core also amounts to 33 GWD/THM. With these models a search for an acceptable core layout is performed:

   - The calculation procedure starts with mean burnup values for all fuel assemblies with the same core residence time. These fuel assemblies must be placed in the core in a way that the overall power distribution is acceptable. The fuel assembly pattern is changed until this requirement is fulfilled.
   - The next step is to find a fuel management scheme. The applied shuffling scheme prescribes the position of every fuel assembly as a function of its reached burnup. These individual burnup stages should be matched to the burnup pattern of the preceding first step as good as possible, taking into account that the core model contains whole and half fuel assemblies. For the central fuel assembly, occupying $\frac{1}{12}$ hexagon, a mean burnup value is taken. The fine tuning of the burnup order of the fuel assemblies in the core is done by test calculations for 1 macro timestep.

6. **Final core simulation.** If a satisfactory core loading and shuffling scheme seems to be found, a final core simulation with several macro timesteps (iterations) follows. In a typical ARCOSI run for a PWR 10 micro timesteps of 30 to 55 equivalent full power days are applied to reach up to 50 GWD/THM mean discharge burnup. Usually one additional macro timestep (iteration) to the number of fuel batches is sufficient to get a stable equilibrium core. The search for the critical boron concentration in the coolant at every micro timestep needs 2 to 3 iterations in most cases. From these considerations follows, that about 100 threedimensional diffusion calculations with 4 energy groups have to be performed for an ARCOSI core simulation run. Every reactor calculation needs the 4 group cross section preparation in dependence of the burnup and the boron concentration in the coolant for 608 separate zones of the applied reactor model. These laborios core simulations could be performed on workstations with the UNIX version of the calculational procedures. The restart options of the program ARCOSI allow further

investigations for the iterated equilibrium core, e.g. additional iterations or calculation of safety related parameters like Doppler coefficients and moderator density reactivity coefficients. The calculation of these safety related parameters need adequate versions of the HXSLIB libraries.

4.2 Results of exploratory whole core calculations.

As a first step to more elaborate long term whole core calculations, a number of exploratory investigations were performed. The primary aim was to get some reliable information about the plutonium fraction needed in the MOX fuel during plutonium multi-recycling in PWRs. In order to validate the applied calculational procedures for these investigations also PWR cores with UOX fuel assemblies were analyzed. In the past a lot of information has been accumulated about PWRs with uranium fuel. Comparison of calculational results with experience from actual reactor operations lead to an acceptable confidence level for the subsequent investigations with plutonium fuel.

The whole core calculations have been started with the analysis of a full MOX core with plutonium from PWRs with 33 GWD/THM discharge burnup, being representative for most of the actually available PWR plutonium. The target burnup of the MOX core is also 33 GWD/THM. For the reactor control only the boration change of the coolant is applied with control rods completely removed from the core. For this case a number of relevant intermediate results are presented in the figures 11 to 16. Figures 11 to 14 show the iteration behaviour and the differences between begin and end of cycle values for the power ratings and the fuel burnup in the core midplane of all fuel assemblies. The latter are represented by their radial distance to the core center. The figures 11 and 13 show a fast convergence for the power rating and the burnup with increasing number of simulations (iterations). Figure 12 shows the radial flattening in the core midplane from begin to end of cycle in the equilibrium core. The corresponding radial power factor decreases from $\approx$1.55 at BOC to $\approx$1.23 at EOC. Figures 15 and 16 give the begin and end of cycle axial distributions of the power rating and the burnup of a selected fuel assembly. The axial power rating also shows a pronounced flattening during the reactor cycle, leading to an improved axial power form factor from $\approx$1.29 at BOC to $\approx$1.08 at EOC. For all performed whole core calculations simular results could be obtained. The main results of the exploratory whole core calculations are summarized in table 5. Information is given about the fuel and its origin, fissile enrichment, desired target burnup, number of fuel batches, cycle length in equivalent full power days and the most important ARCOSI results K_{∞}^{EOC} and mean and maximum discharge burnup.

Case 1 is a calculation for an UOX core with 4% U^{235} enrichment. The resulting mean discharge burnup of $\approx$43 GWD/THM seems not to be in contradiction with actual experiences with discharge burnups in PWRs.

In case 2 plutonium from existing PWRs with 3.2% U^{235} enrichment and 33 GWD/THM mean discharge burnup is investigated. With about 3.5% Pu_{fis} enrichment comparable discharge burnups of about 33 GWD/THM may be obtained.

The cases 3 and 4 give corresponding results for mean target burnups of 40 GWD/THM with plutonium from two generations. The first generation plutonium comes from case 1 (4% U^{235} and 40 GWD/THM mean discharge burnup). In case 3 with this MOX1 enriched at 4.5% Pu_{fis} a mean discharge burnup of 40 GWD/THM can be reached with 3 fuel batches. The most unfavourable use of the second generation plutonium from case 3 is the direct use in a succeeding PWR core without the admixing of better quality plutonium. Case 4 shows, that with this second generation plutonium also 40 GWD/THM mean discharge burnup are obtainable with 6% Pu_{fis} enrichment and the application of 6 fuel batches.

The cases 5 and 6 are calculations for target burnups of 50 GWD/THM. MOX1 comes from PWR UOX fuel with 4.5% U^{235} and 50 GWD/THM discharge burnup. 50 GWD/THM may be obtained with ≈6% Pu_{fis} and 3 fuel batches or with ≈5.5% Pu_{fis} and 6 fuel batches.

In table 6 information about plutonium compositions is summarized. In addition to the cases 2, 3, 4 and 5 from table 5, data from MOX loadings in the nuclear power plant Philippsburg-2 and from benchmark results in the references [4, 5], are provided. Comparing results of the whole core investigation with the benchmark results the different cooling and fabrication times must be kept in mind; the smaller ex-core times of the benchmark lead to less decay of Pu^{241}. Case 5 from table 5 and the first cycle plutonium of the benchmark are comparable; due to the smaller ex-core time the Pu^{241} content of the benchmark plutonium is somewhat higher.

The comparison of the table 5 case 4 plutonium with the benchmark cycle 4 plutonium loading gives some further interesting information:

- The plutonium compositions in these cases are quite similar, so the plutonium from first generation MOX fuel assemblies from PWRs could be used for experiments to validate plutonium behaviour of the fourth plutonium recycling in PWRs.

- The calculated discharge burnup of the whole core calculation with this plutonium, 40 GWD/THM with Pu_{fis}=6%, is comparable with the data of the benchmark, 50 GWD/THM with Pu_{fis} ≈7%. So we have here an indication that the plutonium enrichment results of the benchmark investigations are reasonable and that the equivalency criteria for UOX and MOX fuel assemblies in PWRs cited in reference [11] are conservative with respect to the required fissile enrichments in MOX fuel assemblies.

Case	Fuel	Pluton. origin Enr/BU	Fission fraction (%)	Target burnup (GWD/THM)	Nr. of cycles	Full power days	ARCOSI-Results		
							K_{eff} EOC	burnup (GWD/THM)	
								mean	max.
1	UOX		4.0	40	3	450	1.0071	42.9	49.6
2	MOX1	3.2/33	3.5	33	3	340	0.9995	32.5	38.4
3	MOX1	4.0/40	4.5	40	3	425	0.9986	41.7	49.2
4	MOX2	4.5/40	6.0	40	6	215	1.0006	42.7	49.3
5	MOX1	4.5/50	6.0	50	3	510	0.9995	49.9	58.7
6	MOX1	4.5/50	5.5	50	6	250	0.9981	50.3	56.7

MOX1: first generation plutonium from PWR UOX fuel assemblies.
MOX2: second generation plutonium from PWR MOX1 BE without MOX1 mixing.

Table 5: Summary of results of exploratory PWR whole core calculations.

Case	Pluton. Enr/BU	Contribution in weight percent						
		Pu^{238}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	Am^{241}	Pu_{fis}
2	3.2/33	1.6	59.0	23.1	9.0	5.9	1.4	68.0
KKP2	3.25/33	1.8	59.0	23.0	12.2	4.0	-	71.2
3	4.0/40	2.0	58.5	22.8	9.2	6.1	1.4	67.7
3a	4.0/40	1.6	64.9	25.2	1.3	6.8	0.2	66.2
4	4.5/40	3.3	44.6	29.2	10.9	10.3	1.7	55.5
BM4	6.9/50	4.3	42.4	27.5	12.7	11.8	1.3	55.1
4a	4.5/40	2.8	50.6	33.2	1.6	11.6	0.2	52.2
5	4.5/50	2.8	55.1	23.3	9.7	7.6	1.5	64.8
BM1	5.8/50	2.8	54.0	23.0	12.0	7.0	1.2	66.0

Case 2,3,4,5: from table 5, 7 years cooling , 3 years fabrication time.
Case 3a,4a: as above with 50 years cooling , 3 years fabrication time.
Case KKP2: MOX from the KKW Philippsburg-2
Case BM1: MOX from the EDF/KFK Benchmark for cycle 1
Case BM4: MOX from the EDF/KFK Benchmark for cycle 4

Remark: The EDF/FZK benchmark defines
- 3 years cooling- and 2 years fabrication time and
- 50 GWD/THM target burnup

Table 6: Summary of applied plutonium compositions.

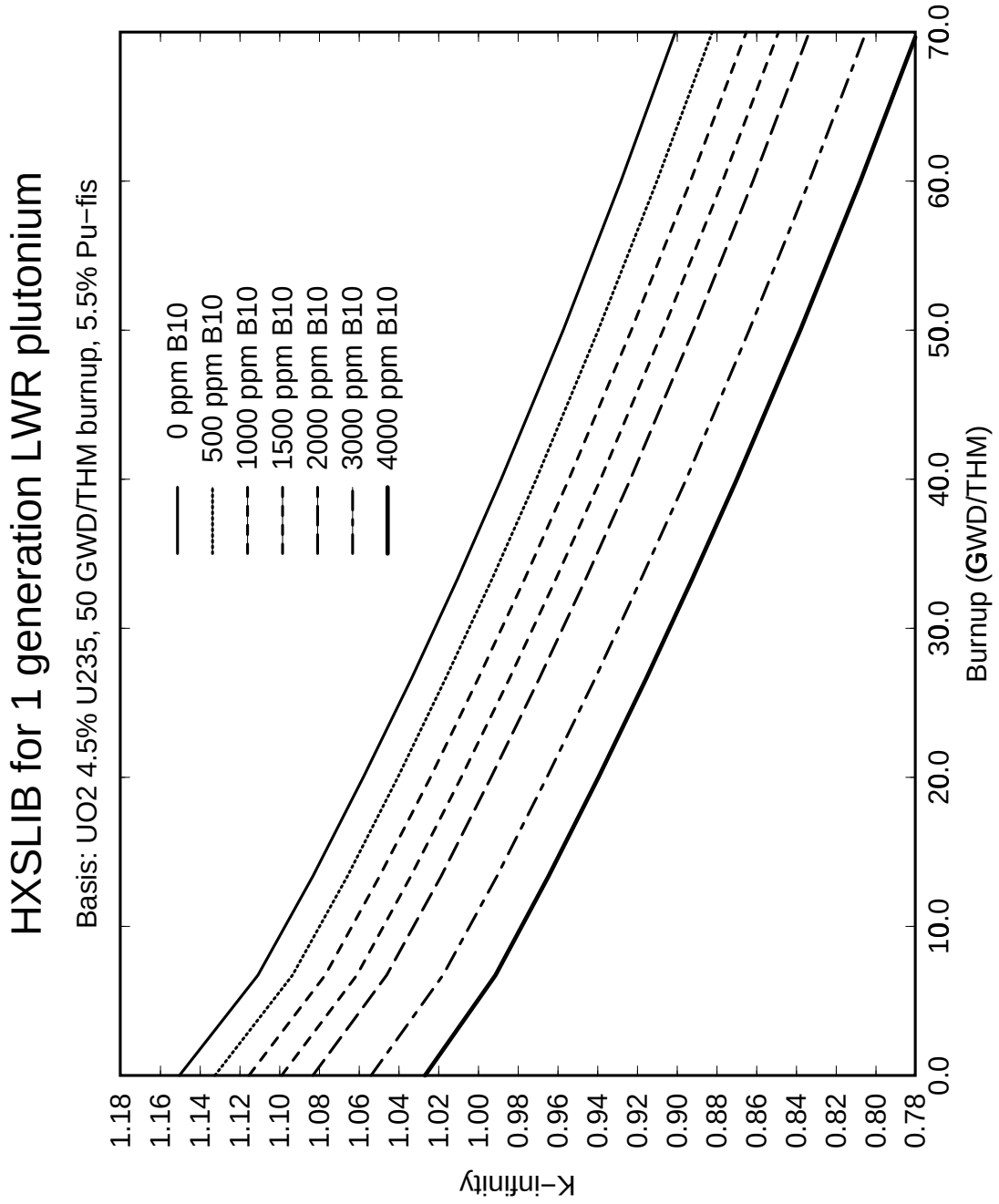

Figure 9: HXSLIB K_∞ data for first generation LWR-MOX.

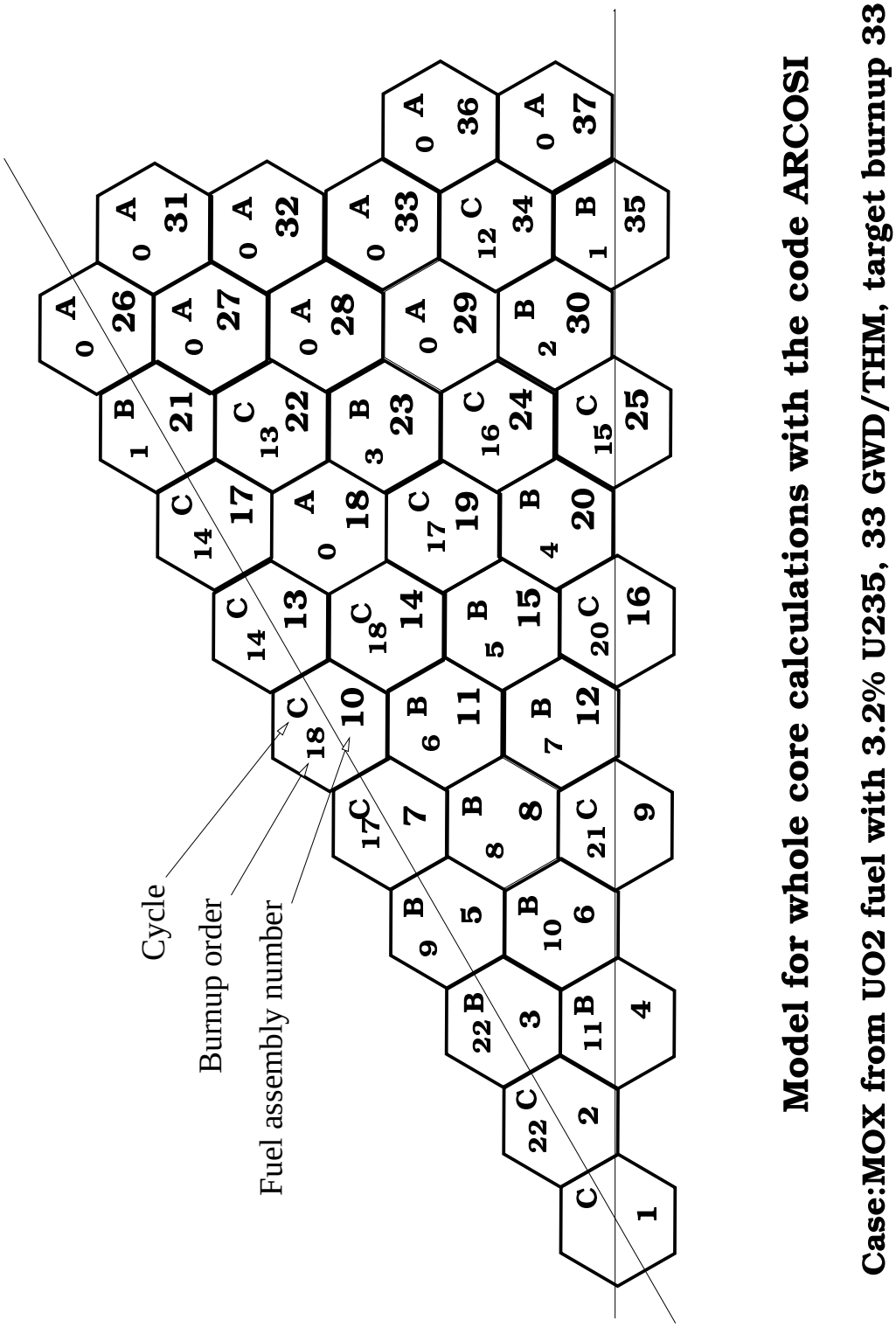

Figure 10: Reactor model for calculations with the code ARCOSI, MOX from UOX fuel, 3.2% U^{235}, 33 GWD/THM discharge burnup, target burnup 33 GWD/THM.

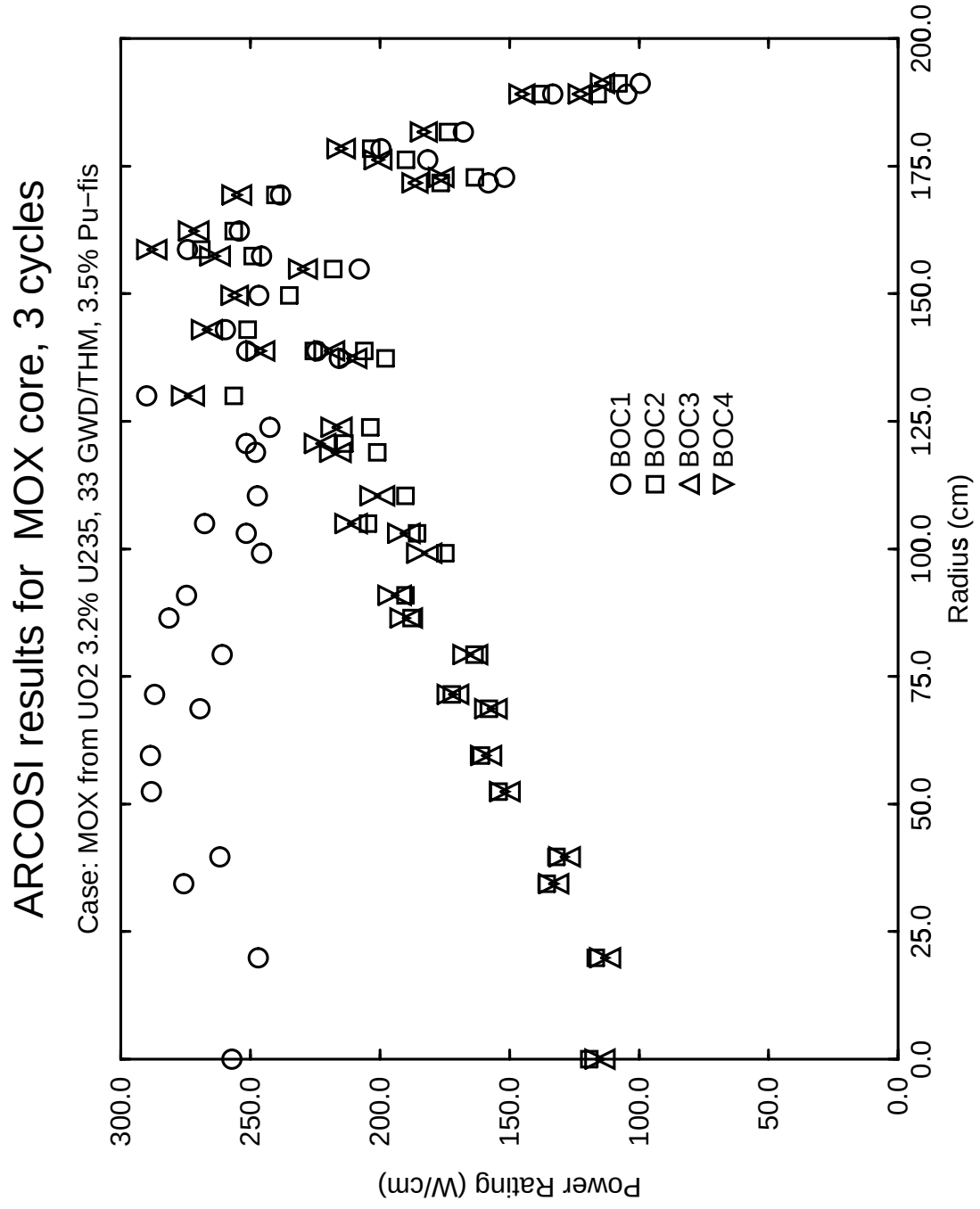

Figure 11: BOC power rating in the axial midplane for 4 succeeding core simulations.

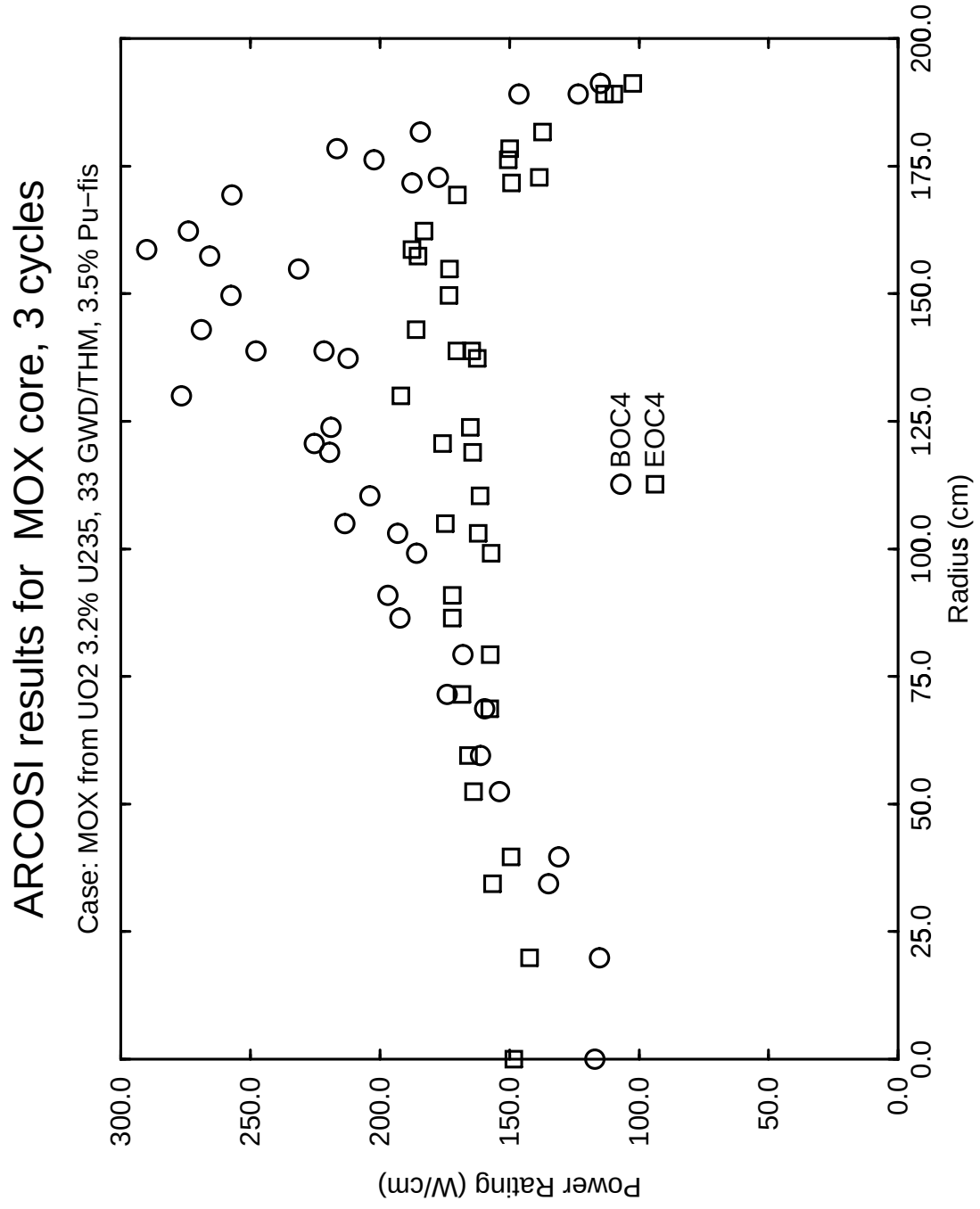

Figure 12: BOC and EOC power ratings in the axial midplane for an equilibrium core.

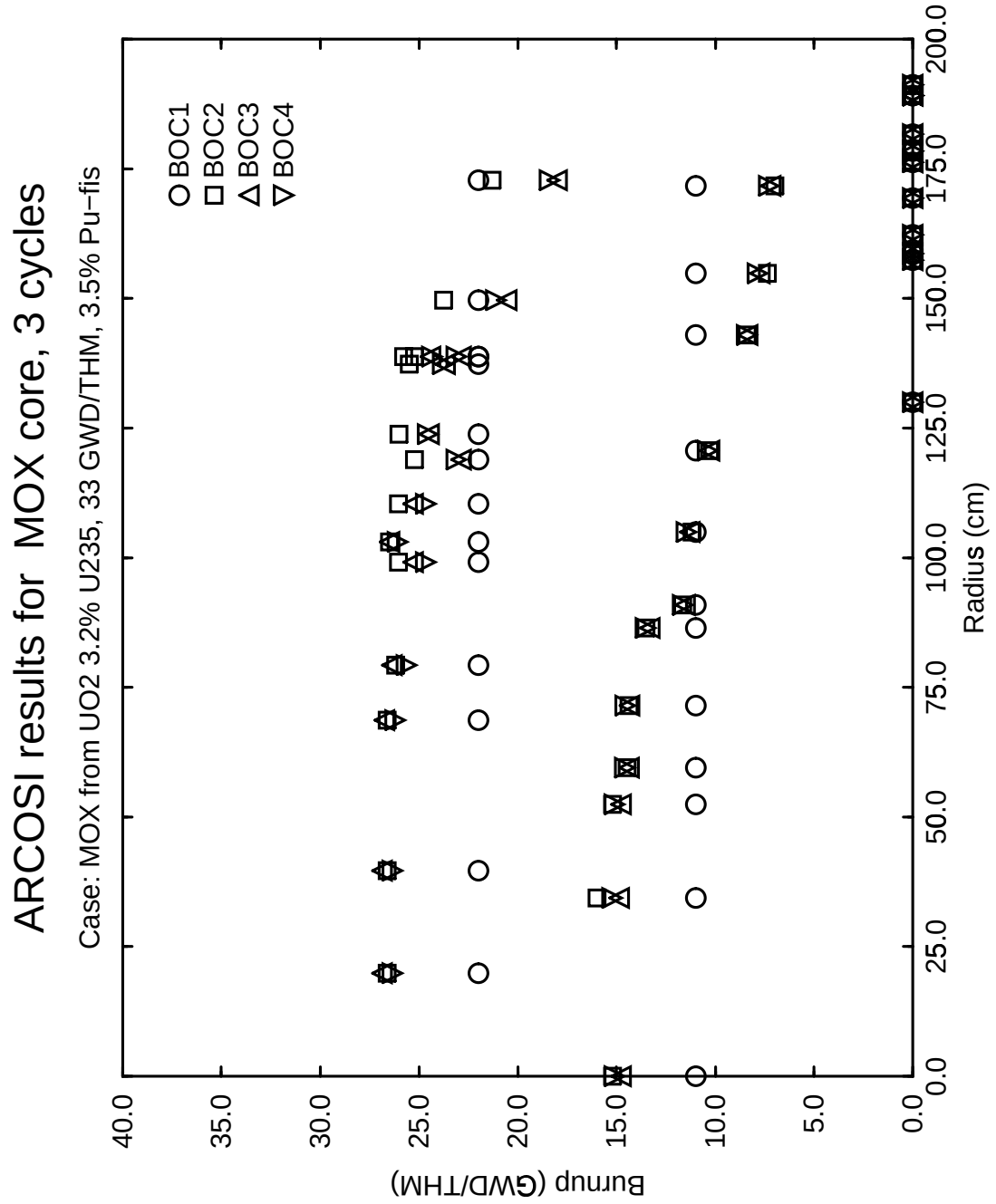

Figure 13: BOC fuel burnup in the axial midplane for 4 succeeding core simulations.

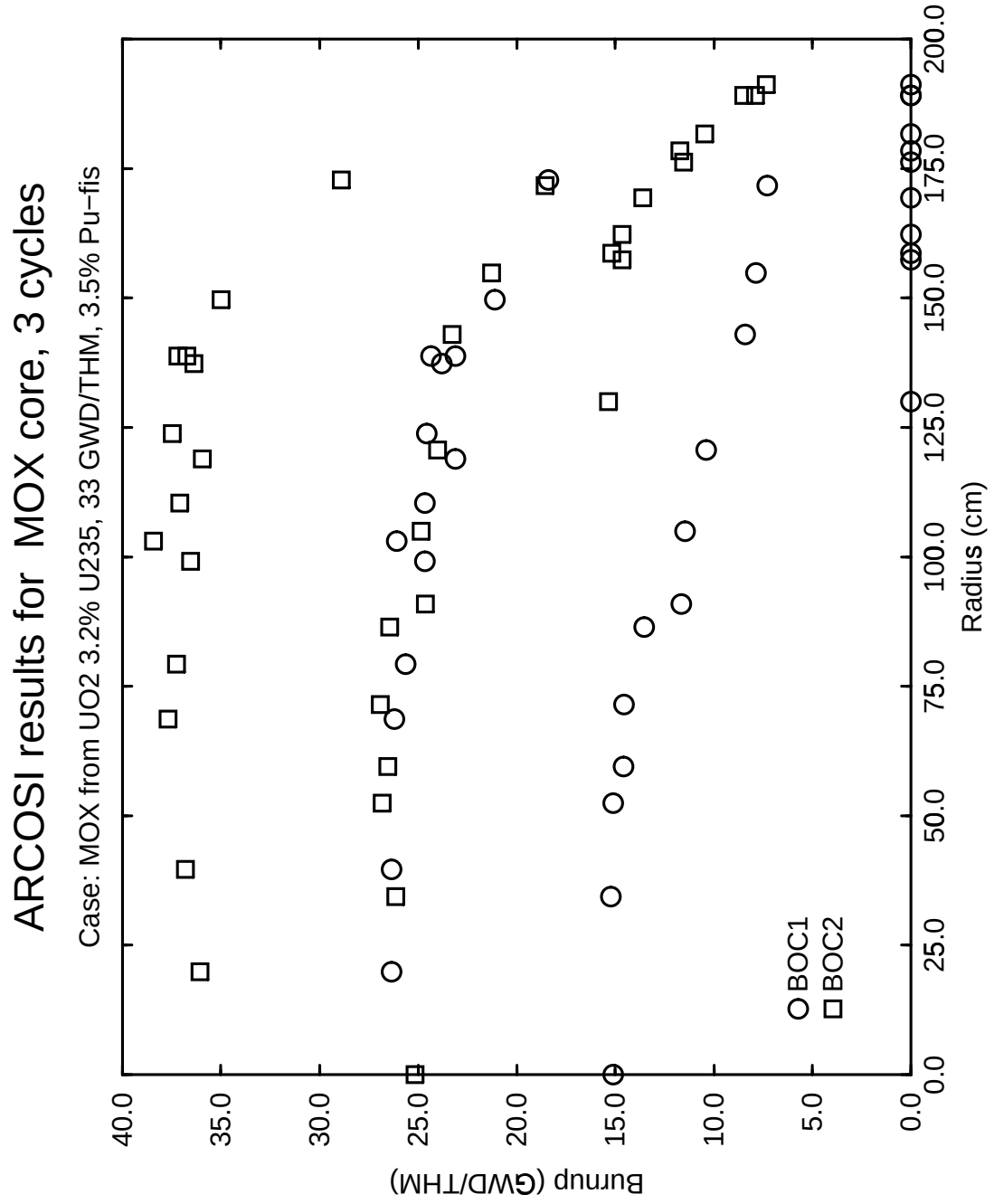

Figure 14: BOC and EOC burnup in the axial midplane of the equilibrium core.

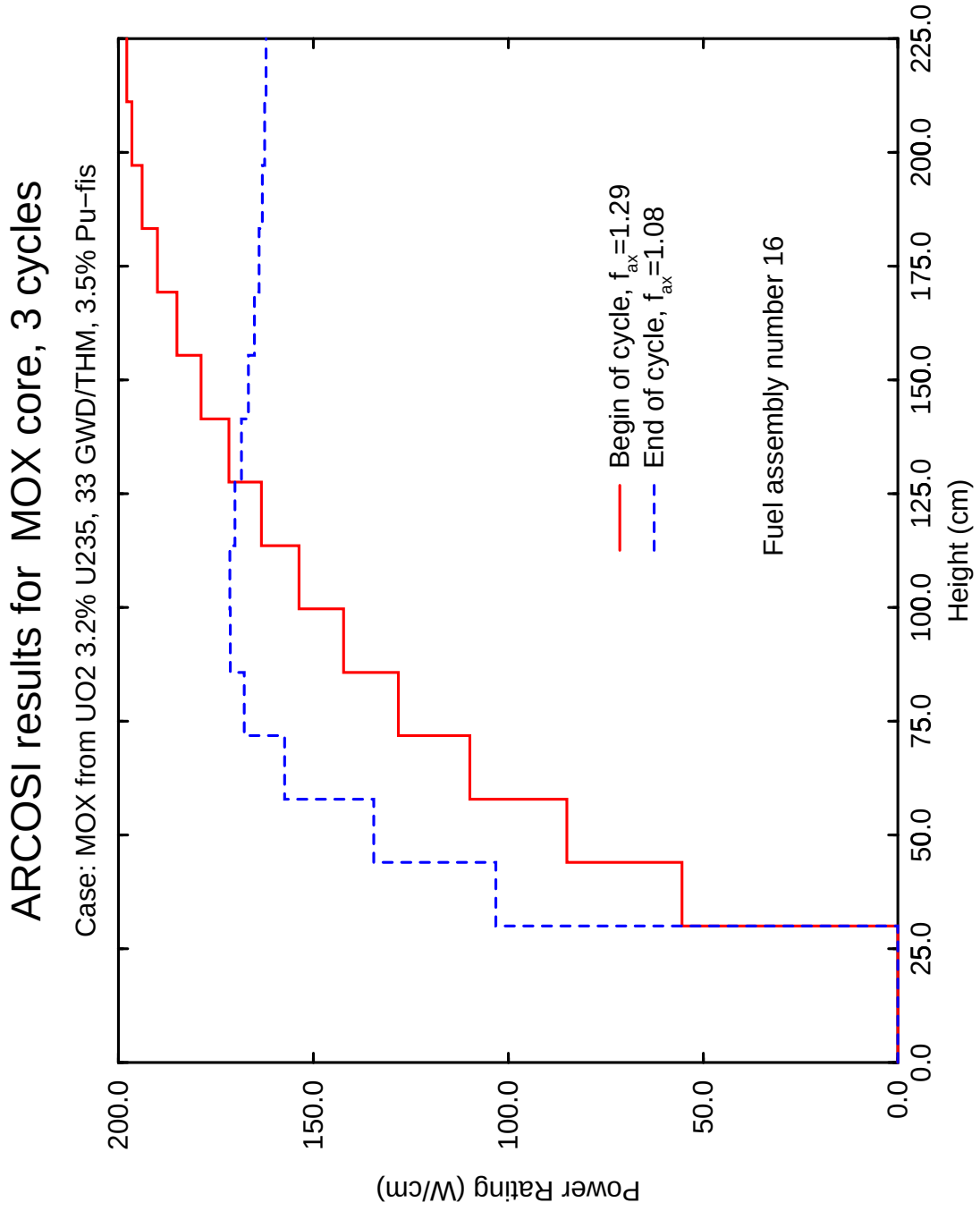

Figure 15: BOC and EOC axial distribution of the power rating of the equilibrium core.

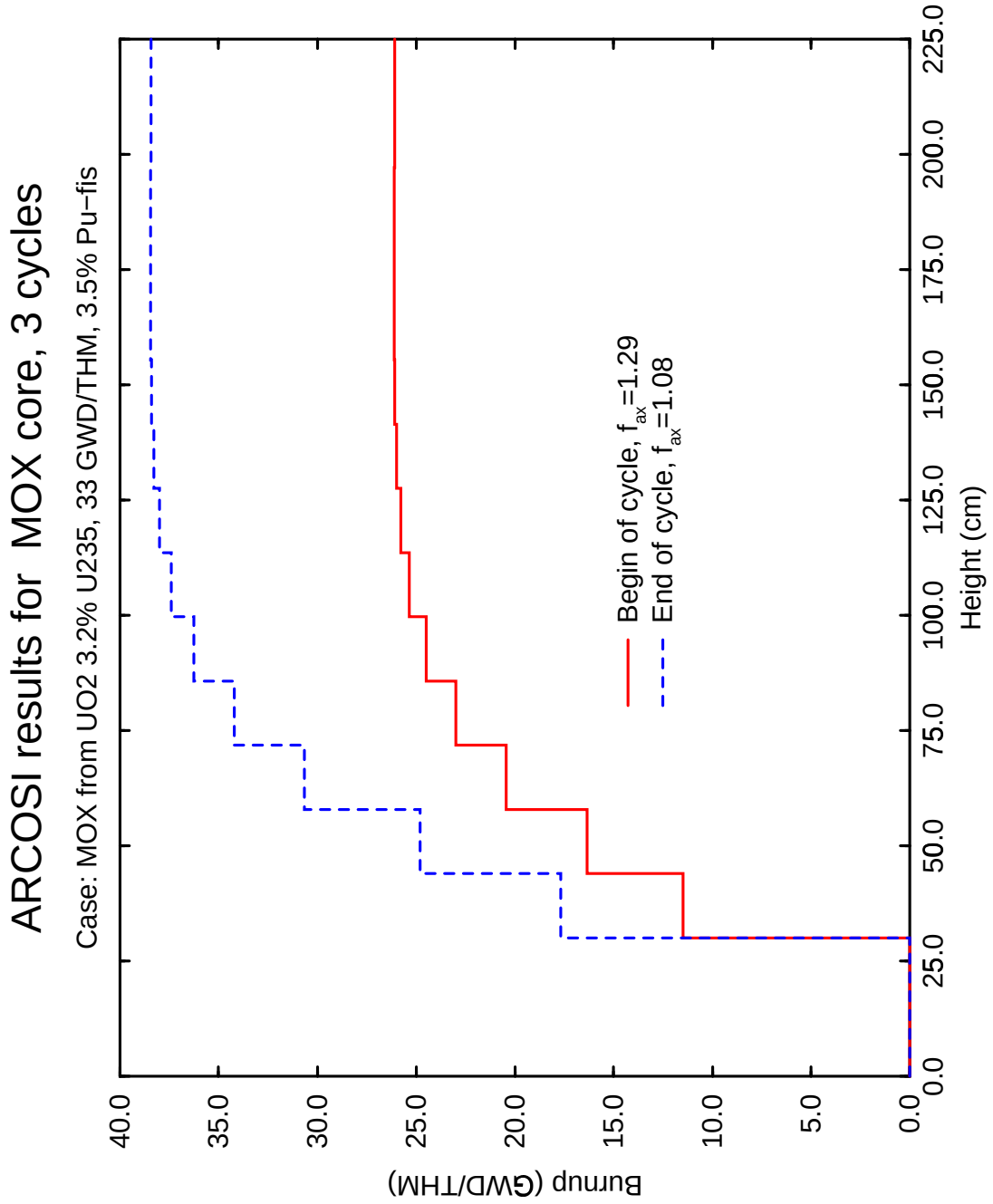

Figure 16: BOC and EOC axial distribution of the fuel burnup of the equilibrium core.

5 Long term considerations for Pu-recycling in PWRs.

One way to reduce the total amount of plutonium produced in PWRs is a decrease of the number of plutonium producing UOX fuel assemblies in such reactors, e.g. by the use of the alternative thorium fuel cycle. Another technically feasible method is the reuse of the produced plutonium in MOX fuel assemblies in PWRs: i.e. plutonium recycling. In the MOX fuel assemblies in PWRs a net incineration of plutonium takes place. The destruction of plutonium by fissions dominates the production by neutron captures in the U^{238}. Furthermore, the neutron capture in the plutonium leads to a shift of the isotopic composition to higher isotopes. The increase of Pu^{240} and Pu^{242} leads to a decrease of the fraction of the fissile isotopes Pu^{239} and Pu^{241}. Moreover the fraction of Pu^{238} increases significantly in MOX fuel assemblies.

5.1 Comparison of plutonium recycling in FBRs and PWRs.

Plutonium recycling is an inherent feature of fast breeder reactors (FBR). So it may be of interest to compare some relevant aspects of plutonium recycling with FBRs and PWRs:

1. Consequences for the fuel fabrication,
2. Consequences for reactor characteristics.

5.1.1 Consequences for the fuel fabrication.

The most important factor during the fabrication of MOX fuel assemblies is the fuel fraction of the isotope Pu^{238} with its hard α-radiation and heat production. According to current knowledge this fraction should not exceed $\approx$10%. In the modern French reprocessing plant MELOX at present the Pu^{238} fraction is limited to $\approx$5% [20]. For this problem no large differences may be expected for MOX fuel of FBRs and PWRs.

Generally the fabrication of MOX fuel assemblies is considerably more expensive compared to UOX fuel assemblies because of the need for remote techniques for the fabrication and handling of MOX fuel.

5.1.2 Consequences for the reactor characteristics.

The consequences of plutonium recycling for the reactor characterisctics, especially the influence on the safety parameters, may be more severe for PWRs compared to FBRs. Whereas FBRs usually have problems with positive coolant density reactivity effects, standard PWRs have sufficiently negative ones. The investigations in reference [1] show, that larger plutonium fractions in PWR MOX fuel assemblies may lead to problems with coolant density reactivity coefficients.

To avoid safety or licensing problems it must be demonstrated, that the loading of MOX fuel assemblies in PWRs does not worsen the safety related reactor parameters, compared to the UOX loadings. Especially the coolant density reactivity coefficients have to remain sufficiently negative. The decrease of the fissile fraction of the plutonium during multi-recycling leads to the need for higher Pu_{fis} fractions and to higher plutonium content in the fuel. Exploratory cell burnup calculations show, that coolant density reactivity effect problems may arise for fuel with Pu_{fis} fractions larger than ≈6..7%. A possible way to achieve the desired target fuel discharge burnups with plutonium multi-recycling in PWRs is to restrict the Pu_{fis} fraction to ≈6% and to use U^{235} enriched uranium in those cases where this is necessary for criticality reasons, see section 4.2.

5.2 Management of the plutonium inventories in the fuel cycle.

Generally it is favourable to have short ex-core times for the plutonium in a closed fuel cycle. The main reason is the relatively short decay time of 14 years of the isotope Pu^{241} with very good fission properties, see for example reference [1]. Further, one has to distinguish between cooling, reprocessing and fuel assembly fabrication times, see section 3.3.1. The succeeding investigations are performed with 7 years (cooling + reprocessing) time and 3 years fabrication time.

The logistic distribution of the available plutonium masses is a very complex problem. Early investigations in this field usually treated the management of the so-called selfgenerated plutonium in PWRs. The EDF/FZK benchmark investigations on plutonium multi-recycling [4, 5], mentioned before, also are based on this model. In the benchmark the MOX content in the PWR cores varies from ≈14% for the first to ≈20..22% for the fifth recycling.

Nowadays the practical application of plutonium recycling in PWRs is based on available plutonium; the self generating aspect is not so important.

Strategic investigations for full MOX PWR cores need new, appropriate solutions for the fuel management scenarios. In the next sections a proposal for such a scenario is presented.

5.3 Use of plutonium in a pool of PWRs.

We consider a pool with a number of identical PWRs with UOX fuel. The reactor cycle time must fulfill the condition that N reactor cycles cover 10 years. These 10 years are chosen to match the ex-core conditions of 7 years (cooling + reprocessing) and 3 years fabrication time (see section 3.3.1). The number of reactors in the pool M is chosen in such a way, that reprocessing of all spent fuel from one cycle of these reactors gives enough plutonium for the startup of one full MOX core. The number of UOX cores then becomes $(M-1)$. At any time all available plutonium from UOX and MOX cores is mixed for the next generation cores. This 10 years scheme is repeated until enough plutonium becomes available to supply another core with MOX fuel. In figure 17 a schematic picture of this scenario is given.

For the fuel inventory calculations a normalisation to 1 ton of initial heavy metal (TIHM) is applied in most cases. The plutonium buildup in a PWR with UOX fuel assemblies varies from ≈9.6 kg/TIHM at 33 GWD/THM to 11.9 kg/TIHM at 50 GWD/THM, see section 3.3.2 table 2.

Rough estimates for the number of UOX cores to supply one MOX core lead to the following results. For a target burnup of 50 GWD/THM in the UOX and MOX fuel a Pu_{fis} enrichment of ≈ 6% is needed with ≈65% fissile fraction in the plutonium. This means a plutonium content of ≈90 kg/TIHM. Table 4 gives 11.9 kg/TIHM for the target burnup of 50 GWD/THM, so 8 UOX fueled PWRs with this discharge burnup are capable to supply 1 MOX core. For the low discharge burnup of 33 GWD/THM these figures are: ≈4% Pu_{fis}, ≈70% fissile fraction in the plutonium and ≈9.6 kg/TIHM plutonium production in the UOX fuel. This means ≈57 kg/THIM are required for a MOX core, equivalent to the production of ≈6 UOX cores.

The pool investigations have started for a mean burnup level of 40 GWD/THM with a pool of 7 PWRs. As a next step the consequences of an increase from 40 to 50 GWD/THM burnup level have been studied. Finally the same investigations have been performed for 33 GWD/THM burnup level, being representative for actually operating PWRs.

5.3.1 Target burnup 40 GWD/THM.

In table 7 a possible scenario for MOX cores in a pool of 7 PWRs with a target burnup of 40 GWD/THM is specified. The UOX cores are identified by the letter U, the MOX cores by M_i for i-th generation plutonium composition. From the 3. recycling, 2 MOX cores within the pool of 7 PWRs are possible. At the 7. recycling, alternatively 2 and 3 MOX cores are considered.

For the scenario of table 7 the following assumptions are made:

1. 4% enrichment of the UOX fuel.
2. 40 GWD/THM target burnup.
3. 7 years (cooling + reprocessing) and 3 years fabrication time.
4. 7 reactor cycles cover 10 years. This means a time between reloadings of 10/7=1.43 years.

The tables 8 to 10 show results for this scenario. Table 8 shows how the different plutonium compositions are build-up. In the first cycles it is taken into account, that not all available plutonium is needed for the MOX core. The remaining part is stored still another 10 years for use in the next generation plutonium. If in the cases with 2 or 3 MOX cores, the available plutonium is not sufficient, it is assumed that the small missing part comes from outside the pool of 7 PWRs and has the same composition of the last generation plutonium. Table 9 summarizes plutonium masses, both for 1 reactor and for the pool of 7 PWRs. The main results are based on burnup per ton of heavy metal. In practice, plutonium and other transurania

masses as a function of the energy production are of interest. In accordance with the ARCOSI calculations the normalized data of table 9 are based on the following assumptions:

- Reactor cycle time T_c from table 10.
- Density of the heavy metal in the fuel rod ρ_f=4.77 g/cm.
- Linear fuel power rating P_l=170 W/cm, being equivalent to a specific fuel power rating of 35.6 Watt/g. In reference [4] a value of 38.3 Watt/g is specified for a 900 MWe French PWR.
- Net electrical power P_E=1300 MWe =$1.3\,10^9$ We.
- Efficiency of the power plant η=0.345.
- Number of fuel cycles N=3.

For the fuel inventory the following relations are valid:

$$G = \frac{P_E \cdot \rho_f}{\eta \cdot P_l} \tag{15}$$

$G = \frac{1.3\,10^9 \cdot 4.77}{0.345 \cdot 170}$ g $= 105.7\,10^6$ g = 105.7 tons.
The discharge fuel per GWe.Year amounts to:

$$G_E = \frac{G \cdot 365}{P_E \cdot T_c \cdot N} \tag{16}$$

Table 9 shows that plutonium multi-recycling in PWRs may lead to a drastical decrease of the plutonium production and even to a net plutonium incineration in the case of 3 MOX cores within a pool of 7 PWRs.

Table 10 summarizes results of whole core calculations for the first 7 plutonium recyclings in the pool of 7 PWRs. Column 4 shows that starting with case 6, enriched U^{235} is required to obtain sufficient criticality with the limited fraction of 6% Pu_{fis} in the MOX. The enrichment varies from 1.2 to 2.2% U^{235} for the target burnup of 40 GWD/THM.

5.3.2 Target burnup 50 GWD/THM.

In table 11 a possible scenario for MOX cores in a pool of 8 PWRs with a target burnup of 50 GWD/THM is specified. From the 3. recycling, 2 MOX cores within the pool of 8 PWRs are possible. At the 8. recycling, 3 MOX cores are considered.

For the scenario of table 11 the following assumptions are made:

1. 4.5% enrichment of the UOX fuel.

2. 50 GWD/THM target burnup.

3. 7 years (cooling + reprocessing) and 3 years fabrication time.

4. 6 reactor cycles cover 10 years. This means a time between reloadings of 10/6=1.67 years.

The tables 12 to 14 show results for this scenario. Table 12 shows how the different plutonium compositions are build-up. Again, in the first cycles it is taken into account, that not all available plutonium is needed for the MOX core. The remaining part is stored still another 10 years for use in the next generation plutonium. If in the cases with 2 or 3 MOX cores, the available plutonium is not sufficient, it is assumed that the small missing part comes from outside the pool of 8 PWRs and has the same composition as that of the last generation plutonium. Table 13 summarizes plutonium masses, both for 1 reactor and for the pool of 8 PWRs. The same assumptions of section 5.3.1 are made.

Table 13 shows that plutonium multi-recycling in PWRs with a target burnup of 50 GWD/THM also may lead to a drastic decrease of the plutonium production and even to a net plutonium incineration in the case of 3 MOX cores within a pool of 8 PWRs.

Table 14 summarizes results of whole core calculations for the first 8 plutonium recyclings in the pool of 7 PWRs. Column 4 shows that starting with case 3, enriched U^{235} is required to obtain sufficient criticality with the limited fraction of 6% Pu_{fis} in the MOX. The enrichment varies from 1.5 to 3.8% U^{235} for the target burnup of 50 GWD/THM.

5.3.3 Target burnup 33 GWD/THM.

For the target burnup of 33 GWD/THM a similar scenario as presented in the tables 7 for 40 GWD/THM and 11 for 50 GWD/THM is applied.

For the scenario for 33 GWD/THM the following assumptions are made:

1. 3.2% enrichment of the UOX fuel.

2. 33 GWD/THM target burnup.

3. 7 years (cooling + reprocessing) and 3 years fabrication time.

4. 8 reactor cycles cover 10 years. This means a time between reloadings of 10/8=1.25 years.

Because of the good experiences with 40 and 50 GWD/THM only a few whole core calculations have been performed. Based on the results of case 2 of table 5 lattice calculations have been performed, using the boundary condition of equation (14).

The tables 15 and 16 show results for this scenario. Table 15 shows how the different plutonium compositions are build-up. Again, in the first cycles it is taken into account, that not all

available plutonium is needed for the MOX core. The remaining part is stored still another 10 years for use in the next generation plutonium. If in the case with 2 MOX cores the available plutonium is not sufficient, it is assumed that the small missing part comes from outside the pool of 6 PWRs and has the same composition of the last generation plutonium. Table 16 summarizes plutonium masses, both for 1 reactor and for the pool of 6 PWRs. The same assumptions of section 5.3.1 are made.

5.3.4 Concluding remarks on plutonium multi-recycling in PWRs.

The preceding investigations for plutonium multi-recycling in a pool of PWRs show some interesting results. The applied constraint of maximum 6% Pu_{fis} in the MOX fuel to avoid problems with coolant density reactivity coefficients leads to the need for a target burnup dependent U^{235} enrichment of the uranium in the MOX fuel. Table 17 gives a summary of the main results for the near equilibrium reactor cycles with target burnups 33, 40 and 50 GWD/THM. We may observe, that the ratio between the plutonium production in the UOX cores and the plutonium incineration in the MOX cores is nearly constant $\approx$0.6. This means that if one has a pool of equal PWRs with a ratio of MOX to UOX cores of 3 to 5, the amount of plutonium in this pool is nearly constant after the near equilibrium reactor cycle is reached. This property is practically independent of the target burnup. However, the required U^{235} enrichment strongly depends on the target burnup, varying from small values for 33 GWD/THM to 4% for 50 GWD/THM.

Time year	Cycle	Reactors						
		1	2	3	4	5	6	7
0	1	U	U	U	U	U	U	U
	2	U	U	U	U	U	U	U
	.	U	U	U	U	U	U	U
10	7	U	U	U	U	U	U	U
	8	U	U	U	U	U	U	M1
	.	U	U	U	U	U	U	M1
20	14	U	U	U	U	U	U	M1
	15	U	U	U	U	U	U	M2
	.	U	U	U	U	U	U	M2
30	21	U	U	U	U	U	U	M2
	22	U	U	U	U	U	M3	M3
	.	U	U	U	U	U	M3	M3
40	28	U	U	U	U	U	M3	M3
	29	U	U	U	U	U	M4	M4
	.	U	U	U	U	U	M4	M4
50	35	U	U	U	U	U	M4	M4
	36	U	U	U	U	U	M5	M5
	.	U	U	U	U	U	M5	M5
60	42	U	U	U	U	U	M5	M5
	43	U	U	U	U	U/M6M	M6/M6M	M6/M6M
	.	U	U	U	U	U/M6M	M6/M6M	M6/M6M
70	49	U	U	U	U	U/M6M	M6/M6M	M6/M6M
	50	U	U	U	U	M7	M7	M7
	.	U	U	U	U	M7	M7	M7
80	56	U	U	U	U	M7	M7	M7

U : UOX
Mi/MiM: MOX generation i

Assumptions: 7 years cooling + 3 years fabrication time, 7 reactor cycles in 10 years.

Table 7: Scenario for the use of full MOX cores in a pool of 7 PWRs, target burnup 40 GWD/THM.

Case	Origin	Fraction in weight%						
		Pu^{238}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	Am^{241}	Pu_{fis}
M1	UOX BE	2.0	58.5	22.8	9.2	6.1	1.4	67.7
M2	6x10.6 kg M1	2.0	58.5	22.8	9.2	6.1	1.4	67.7
	8.4 kg $M1'$	1.9	61.1	23.8	5.9	6.4	0.9	67.0
	51.1 kg M2	3.3	44.0	29.8	10.8	10.4	1.7	55.8
	Mixture 123.1 kg	2.5	52.7	25.8	9.6	7.9	1.5	62.3
M3	6x10.6 kg M1	2.0	58.5	22.8	9.2	6.1	1.4	67.7
	43.7 kg $M2'$	3.3	46.3	31.4	7.0	10.9	1.1	53.3
	63.2 kg M3	3.7	41.7	30.7	10.6	11.7	1.6	52.3
	Mixture 170.5 kg	3.0	49.1	27.9	9.2	9.4	1.4	58.3
M4	5x10.6 kg M1	2.0	58.5	22.8	9.2	6.1	1.4	67.7
	2x75.9 kg M4	4.0	40.0	31.6	10.1	12.7	1.6	50.1
	Mixture 204.8 kg	3.5	44.8	29.3	9.9	11.0	1.5	54.7
M5	5x10.6 kg M1	2.0	58.5	22.8	9.2	6.1	1.4	67.7
	2x92.9 kg M4	4.4	38.0	32.2	9.8	14.1	1.5	47.8
	Mixture 238.5 kg	3.9	42.5	30.1	9.7	12.3	1.5	52.2
M6	5x10.6 kg M1	2.0	58.5	22.8	9.2	6.1	1.4	67.7
	2x95.2 kg M5	4.7	36.5	32.3	9.6	15.4	1.5	46.1
	Mixture 243.4 kg	4.1	41.3	30.2	9.5	13.3	1.5	50.8
M6M	4x10.6 kg M1	2.0	58.5	22.8	9.2	6.1	1.4	67.7
	3x95.2 kg M5	4.7	36.5	32.3	9.6	15.4	1.5	46.1
	Mixture 328.0 kg	4.3	39.3	31.1	9.6	14.2	1.5	48.9
M7	4x10.6 kg M1	2.0	58.5	22.8	9.2	6.1	1.4	67.7
	3x94.4 kg M6M	4.8	35.8	32.1	9.4	16.4	1.5	45.2
	Mixture 325.6 kg	4.4	38.8	30.9	9.4	15.0	1.5	48.2

M1,M2...M6M: 7 years cooling and 3 years fabrication time
$M1'$,$M2'$: 17 years cooling and 3 years fabrication time

Table 8: Summary of plutonium mixing for target burnups of 40 GWD/THM.

Case	Reactor	Plutonium in kg/TIHM				Plutonium in kg/GWeYear		
		Reactor		Balance				
		Input	Output	Reactor	Pool	Reactor	Pool	Mean
1	U	-	10.6	+10.6	+74.2	+233.4	+1632.7	+233.4
2	M1	65.8	51.1	-14.7	+48.9	-342.5	+1139.3	+162.8
3	M2	79.4	63.2	-16.2	+47.4	-401.0	+1173.4	+167.6
4	M3	93.5	75.9	-17.6	+46.0	-435.7	+1138.7	+162.7
5	M4	112.3	92.9	-19.4	+14.2	-480.2	+351.5	+50.2
6	M5	113.9	95.2	-18.7	+15.6	-462.9	+386.2	+55.2
7	M6	117.0	98.5	-18.5	+16.0	-458.0	+396.1	+56.6
8	M6M1	121.6	103.2	-18.4	-12.8	-455.5	-316.9	-45.3
9	M6M2	111.4	94.4	-17.0	-8.6	-420.8	-212.9	-30.4
10	M7	123.4	104.9	-18.5	-13.1	-458.0	-324.3	-46.3

Identifications U,M1,M2..M7 from table 8.

Table 9: Plutonium masses in a pool of 7 PWRs with target burnups of 40 GWD/THM.

Case	Fuel	Pluton. origin Enr/BU	Fiss. fr. (%) Pu/U	Target-BU (GWD/THM)	Nr. Cyc-les	Cycl. time (fpd)	ARCOSI results		
							K_{eff} EOC	Burnup (GWD/THM)	
								Mean	Max.
1	UOX		0.0/4.0	40	3	450	1.0071	42.9	49.6
2	M1	4.0/40	4.5/0.7	40	3	425	0.9986	41.7	49.2
3	M2	4.5/40	5.0/0.7	40	3	400	1.0012	39.4	48.2
4	M3	5.0/40	5.5/0.7	40	3	400	0.9985	39.4	49.4
5	M4	5.5/40	6.2/0.7	40	3	400	0.9992	39.4	50.2
6	M5	6.2/40	6.0/1.2	40	3	400	0.9976	39.4	50.6
7	M6	6.0/40	6.0/1.5	40	3	400	0.9984	39.5	50.9
8	M6M1	6.0/40	6.0/1.9	40	3	400	0.9997	39.5	51.2
9	M6M2	6.0/40	5.5/2.2	40	3	400	0.9989	39.5	51.1
10	M7	6.0/40	6.0/2.0	40	3	400	0.9990	39.5	51.3

Table 10: Whole core calculations for a pool of 7 PWRs with target burnups of 40 GWD/THM.

Time year	Cycle	Reactors							
		1	2	3	4	5	6	7	8
0	1	U	U	U	U	U	U	U	U
	2	U	U	U	U	U	U	U	U
	.	U	U	U	U	U	U	U	U
10	6	U	U	U	U	U	U	U	U
	7	U	U	U	U	U	U	U	M1
	.	U	U	U	U	U	U	U	M1
20	12	U	U	U	U	U	U	U	M1
	13	U	U	U	U	U	U	U	M2
	.	U	U	U	U	U	U	U	M2
30	18	U	U	U	U	U	U	U	M2
	19	U	U	U	U	U	U	M3	M3
	.	U	U	U	U	U	U	M3	M3
40	24	U	U	U	U	U	U	M3	M3
	25	U	U	U	U	U	U	M4	M4
	.	U	U	U	U	U	U	M4	M4
50	30	U	U	U	U	U	U	M4	M4
	31	U	U	U	U	U	U	M5	M5
	.	U	U	U	U	U	U	M5	M5
60	36	U	U	U	U	U	U	M5	M5
	37	U	U	U	U	U	U	M6	M6
	.	U	U	U	U	U	U	M6	M6
70	42	U	U	U	U	U	U	M6	M6
	43	U	U	U	U	U	U	M7	M7
	.	U	U	U	U	U	U	M7	M7
80	48	U	U	U	U	U	U	M7	M7
	49	U	U	U	U	U	M8	M8	M8
	.	U	U	U	U	U	M8	M8	M8
90	54	U	U	U	U	U	M8	M8	M8

U : UOX
Mi: MOX generation i

Assumptions: 7 years cooling + 4 years fabrication time, 6 cycles in 10 years.

Table 11: Scenario for the use of full MOX cores in a pool of 8 PWRs, target burnups 50 GWD/THM.

Case	Origin	Fraction in weight%						
		Pu^{238}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	Am^{241}	Pu_{fis}
M1	UOX BE	2.8	55.1	23.3	9.7	7.6	1.5	64.8
M2	7x11.9 kg M1	2.8	55.1	23.3	9.7	7.6	1.5	64.8
	3.6 kg $M1'$	2.7	57.7	24.4	6.3	7.9	1.0	64.0
	69.8 kg M2	4.3	42.2	29.7	10.5	11.7	1.6	52.7
	Mixture 156.7 kg	3.5	49.4	26.2	10.0	9.4	1.5	59.4
M3	7x11.9 kg M1	2.8	55.1	23.3	9.7	7.6	1.5	64.8
	56.0 kg $M2'$	4.2	44.4	31.2	6.8	12.3	1.1	51.2
	78.6 kg M3	4.7	39.8	30.4	10.3	13.2	1.6	50.1
	Mixture 217.9 kg	3.9	46.8	27.9	9.2	10.8	1.4	56.0
M4	6x11.9 kg M1	2.8	55.1	23.3	9.7	7.6	1.5	64.8
	5.4 kg $M3'$	4.7	41.8	31.9	6.7	13.9	1.0	48.5
	2x89.0 kg M4	4.9	38.3	31.0	10.0	14.2	1.6	48.3
	Mixture 254.8 kg	4.3	43.1	28.9	9.9	12.3	1.5	53.0
M5	6x11.9 kg M1	2.8	55.1	23.3	9.7	7.6	1.5	64.8
	30.4 kg $M4'$	4.8	40.2	32.6	6.5	14.9	1.0	46.7
	2x91.0 kg M5	5.3	36.4	31.1	9.9	15.8	1.5	46.3
	Mixture 283.8 kg	4.6	41.5	29.3	9.5	13.6	1.5	51.0
M6	6x11.9 kg M1	2.8	55.1	23.3	9.7	7.6	1.5	64.8
	50.6 kg $M5'$	5.1	38.3	32.7	6.4	16.5	1.0	44.7
	2x95.7 kg M6	5.4	35.5	31.1	9.6	16.9	1.5	45.1
	Mixture 313.4 kg	4.8	40.4	29.6	9.1	14.7	1.4	49.5
M7	6x11.9 kg M1	2.8	55.1	23.3	9.7	7.6	1.5	64.8
	66.6 kg $M6'$	5.3	37.2	32.6	6.2	17.7	1.0	43.4
	2x100.6 kg M6	5.5	34.8	31.1	9.3	17.8	1.5	44.1
	Mixture 339.2 kg	4.9	39.5	29.8	8.8	15.6	1.4	48.3
M8	5x11.9 kg M1	2.8	55.1	23.3	9.7	7.6	1.5	64.8
	3x102.3 kg M7	5.5	34.1	31.1	9.2	18.7	1.4	43.3
	Mixture 366.4 kg	5.1	37.5	29.8	9.3	16.9	1.4	46.8

M1,M2...M8: 7 years cooling and 3 years fabrication time
$M1'$,$M6'$: 17 years cooling and 3 years fabrication time

Table 12: Summary of plutonium mixing for target burnups of 50 GWD/THM.

Case	Reactor	Plutonium in kg/TIHM				Plutonium in kg/GWeYear		
		Pro reactor		Balance				
		Input	Output	Reactor	Pool	Reactor	Pool	Mean
1	U	-	11.9	+11.9	+95.2	+218.2	+1745.6	+218.2
2	M1	91.6	69.8	-21.8	+61.5	-423.3	+1194.0	+149.3
3	M2	100.0	78.6	-21.4	+61.9	-415.5	+1201.8	+150.2
4	M3	106.2	85.0	-21.2	+62.1	-411.6	+1205.7	+150.7
5	M4	112.2	91.0	-21.2	+29.0	-411.6	+563.0	+70.4
6	M5	116.6	95.7	-20.9	+29.6	-405.8	+574.7	+71.8
7	M6	120.3	99.4	-20.9	+29.6	-405.8	+574.7	+71.8
8	M7	123.3	102.3	-21.0	+29.4	-407.7	+570.8	+71.4
9	M8	127.2	106.2	-21.0	-3.5	-407.7	-68.0	-8.5

Identifications U,M1,M2..M8 from table 13.

Table 13: Plutonium masses in a pool of 8 PWRs with target burnups of 50 GWD/THM.

Case	Fuel	Pluton. origin Enr/BU	Fiss. fr. (%) Pu/U	Target-BU (GWD/THM)	Nr. Cyc-les	Cycl. time (fpd)	ARCOSI results		
							K_{eff} EOC	Burnup (GWD/THM)	
								Mean	Max.
1	UOX		0.0/4.5	50	3	540	0.9997	50.5	58.8
2	M1	4.5/50	6.0/0.7	50	3	510	0.9995	49.9	58.7
3	M2	6.0/50	6.0/1.5	50	3	510	1.0013	50.0	59.5
4	M3	6.0/50	6.0/2.0	50	3	510	1.0010	50.1	60.9
5	M4	6.0/50	6.0/2.5	50	3	510	1.0017	50.1	61.1
6	M5	6.0/50	6.0/3.0	50	3	510	1.0040	50.1	61.5
7	M6	6.0/50	6.0/3.3	50	3	510	1.0044	50.1	61.8
8	M7	6.0/50	6.0/3.5	50	3	510	1.0039	50.1	62.0
9	M8	6.0/50	6.0/3.8	50	3	510	1.0047	50.1	62.0

Table 14: Whole core calculations for a pool of 8 PWRs with target burnups of 50 GWD/THM.

Case	Origin	Fraction in weight%						
		Pu^{238}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	Am^{241}	Pu_{fis}
M1	UOX BE	1.6	59.0	23.1	9.0	5.9	1.4	68.0
M2	5x9.6 kg M1	1.6	59.0	23.1	9.0	5.9	1.4	68.0
	6.7 kg $M1'$	1.5	61.6	24.1	5.8	6.1	0.9	67.4
	40.5 kg M2	2.8	44.2	30.0	10.9	10.4	1.7	55.1
	Mixture 95.2 kg	2.1	52.9	26.1	9.6	7.8	1.5	62.5
M3	5x9.6 kg M1	1.6	59.0	23.1	9.0	5.9	1.4	68.0
	31.9 kg $M2'$	2.8	46.5	31.6	7.1	10.9	1.1	53.6
	51.4 kg M3	3.2	42.0	30.7	10.7	11.7	1.7	52.7
	Mixture 131.3 kg	2.5	49.3	28.1	9.2	9.4	1.5	58.5
M4	5x9.6 kg M1	1.6	59.0	23.1	9.0	5.9	1.4	68.0
	55.2 kg M4	3.1	44.2	32.3	7.0	12.3	1.1	51.2
	62.9 kg M3	3.4	40.4	31.6	10.3	12.7	1.6	50.7
	Mixture 166.1 kg	2.8	47.0	29.4	8.8	10.6	1.4	55.8
M5	4x9.6 kg M1	1.6	59.0	23.1	9.0	5.9	1.4	68.0
	2x74.5 kg M4	3.6	39.3	32.3	9.8	13.5	1.5	49.1
	Mixture 187.4 kg	3.2	43.3	30.4	9.6	11.9	1.5	52.9
M6	4x9.6 kg M1	1.6	59.0	23.1	9.0	5.9	1.4	68.0
	2x90.8 kg M5	3.9	37.7	32.7	9.5	14.7	1.5	47.2
	Mixture 220.0 kg	3.5	41.4	31.0	9.4	13.2	1.5	50.8
M7	4x9.6 kg M1	1.6	59.0	23.1	9.0	5.9	1.4	68.0
	2x100.8 kg M6	4.1	36.5	33.0	9.2	15.8	1.4	45.7
	Mixture 240.0 kg	3.7	40.1	31.4	9.2	14.2	1.4	49.3

M1,M2...M7: 7 years cooling and 3 years fabrication time
$M1'$,$M2'$: 17 years cooling and 3 years fabrication time

Table 15: Summary of plutonium mixing for target burnups of 33 GWD/THM.

Case	Reactor	Plutonium in kg/TIHM				Plutonium in kg/GWeYear		
		Pro reactor		Balance				
		Input	Output	Reactor	Pool	Reactor	Pool	Mean
1	U	-	9.6	+9.6	+57.6	+260.2	+1561.2	+260.2
2	M1	50.9	40.5	-10.4	+37.6	-281.9	+1019.1	+169.9
3	M2	63.3	51.4	-11.9	+36.1	-322.5	+978.4	+163.1
4	M3	88.8	74.5	-13.2	+33.7	-387.6	+913.4	+152.2
5	M4	106.5	90.8	-15.7	+22.7	-425.5	+615.2	+102.5
6	M5	117.0	100.8	-16.2	+22.2	-439.1	+601.7	+100.3
7	M6	120.8	104.4	-16.4	+22.0	-444.5	+596.3	+99.4

Identifications U,M1,M2..M6 like table 8.

Table 16: Plutonium masses in a pool of 6 PWRs with target burnups of 33 GWD/THM.

Target-burnup (GWD/THM)	Pu-balance (Kg/TIHM)		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 17: Results for near equilibrium cycles in pools of PWRs with UOX and MOX fuel.

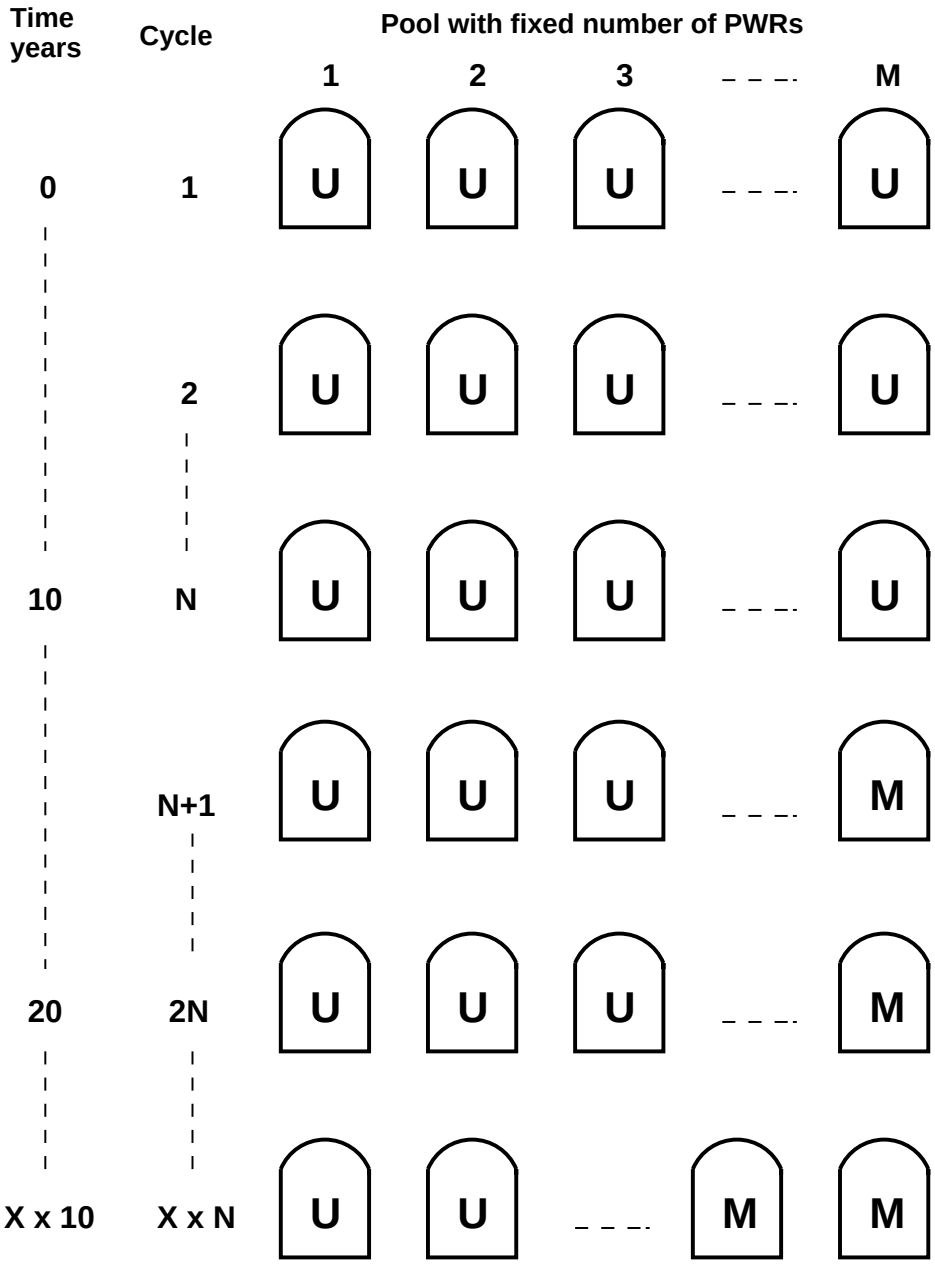

Figure 17: Scenario for plutonium multi-recycling in a pool of PWRs.

6 Plutonium recycling and safety related parameters.

From earlier investigations, e.g. in reference [1], it is well-known that the plutonium partition in the MOX fuel of PWRs is limited if safety margins have to be preserved. The most important reason for this limitation is the behaviour of the coolant density reactivity coefficient, see section 5.1.2. The second important safety parameter, the prompt fuel temperature reactivity coefficient (Doppler effect), is less sensitive to the MOX content and to the layout of the fuel, see e.g. chapter 6.1.3 in reference [1]. In the next sections some investigations related to the coolant density reactivity coefficients are presented. Both lattice and whole core calculations are performed.

6.1 Coolant density reactivity coefficients of fuel lattices.

A first indication for the coolant density reactivity coefficients of a reactor system is the reactivity change due to the total removal of the coolant from the basic fuel lattice (void effect).

The void reactivity

$$\Delta K_{\infty,Void} = K_{\infty,Void} - K_{\infty,Normal} \tag{17}$$

must be sufficiently negative. Otherwise more accurate whole core calculations are required.

The figures 18 and 19 show K_∞ curves as a function of fuel burnup for 2. and 3. generation plutonium from discharge burnups of 50 GWD/THM. Starting from UOX lattices with 4.5% U^{235} enrichment and 50 GWD/THM discharge burnup the 1. generation plutonium is mixed with natural uranium to MOX1. The MOX1 is irradiated up to 50 GWD/THM burnup, using 5% Pu_{fis} enrichment. The produced 2. generation plutonium is used directly in the next core, without blending it with better quality plutonium. In this case 5.8% Pu_{fis} enrichment with natural uranium gives MOX2. The irradiation of MOX2 up to 50 GWD/THM leads to the bad quality 3. generation plutonium. For this 3. generation plutonium burnup calculations have been performed for 2 fuel composotions: 7.5% Pu_{fis} with natural uranium and 6% $Pufis$ with 2% U^{235} enriched uranium.

In the figure 18 we may observe, that for the 2. generation plutonium with 6% Pu_{fis} no problems arise with respect to coolant density reactivity coefficients.

For the 3. generation plutonium figure 19 shows large positive void effects for the case natural uranium/7.5% Pu_{fis}. The use of 2% U^{235} enriched uranium enables a decrease to 6% Pu_{fis}, leading to near zero void effects. For such fuel, more accurate whole core calculations are required to get sufficient knowledge about the coolant density reactivity coefficients of the reactor.

For the cases 8, 9 and 10 from the tables 9 and 10 and for case 8 from the tables 14 and 15 first void effect investigations were performed. In figure 20 the K_∞ curves for the M6M

lattices with 5.5/6.0% Pu_{fis} and 2.2/1.9% U^{235} are plotted. Figure 21 shows K_∞ curves for higher cycles of plutonium multi-recycling with discharge burnup of 40 and 50 GWD/THM. These results are not sufficient to ensure that the coolant density reactivity coefficients have no impact on safety aspects of the PWR. This question only may be clarified by whole core investigations.

6.2 Whole core coolant density reactivity coefficients.

Due to the leakage from the core, in most cases the coolant density reactivity coefficients in a real core are more favourable as in the basic lattices. In a first approximation the leakage may be taken into account in lattice calculations by means of so-called bucklings, e.g. the geometrical buckling defined by the geometry of the reactor. The leakage in the harder neutron spectrum of the voided lattice is larger than in the normal lattice, leading to a negative additional component to the void reactivity. A further effect comes from the very heterogeous burnup distribution in the core with a tendency to higher burnups in the center, see e.g. figure 16. The smaller burnup near the core boundary leads to an increase of the leakage, compared to a homogeneous core.

For the cases of section 6.1 therefore whole core calculations have been performed with modified coolant densities. Special HXSLIB libraries with 50% of the nominal coolant density have been prepared for use with the code ARCOSI. With the restart options of ARCOSI, whole core calculations may be performed for selected cases of the equilibrium cores. Begin of life with maximum amount of boron in the coolant and the boron-free end of life situation have been chosen. Figure 22 shows the results of these calculations. The curves are identified by numbers, representing Pu_{fis} and U^{235} enrichment, number of the plutonium recycling, target burnup and B^{10} content in ppm in the coolant. All coolant density decreases lead to clearly negative reactivity effects. The begin of cycle conditions are more unfavourable because of the loss of boron together with the coolant. For higher target burnups the needed boron concentration increases with unfavourable consequences for the reactivity changes. However, this disadvantage might be reduced by use of banks of control rods inserted for the begin of life condition.

The results of reference [1] indicate, that coolant density reactivity curves as found in figure 22 are satisfactory for the replacement of the core of modern PWRs.

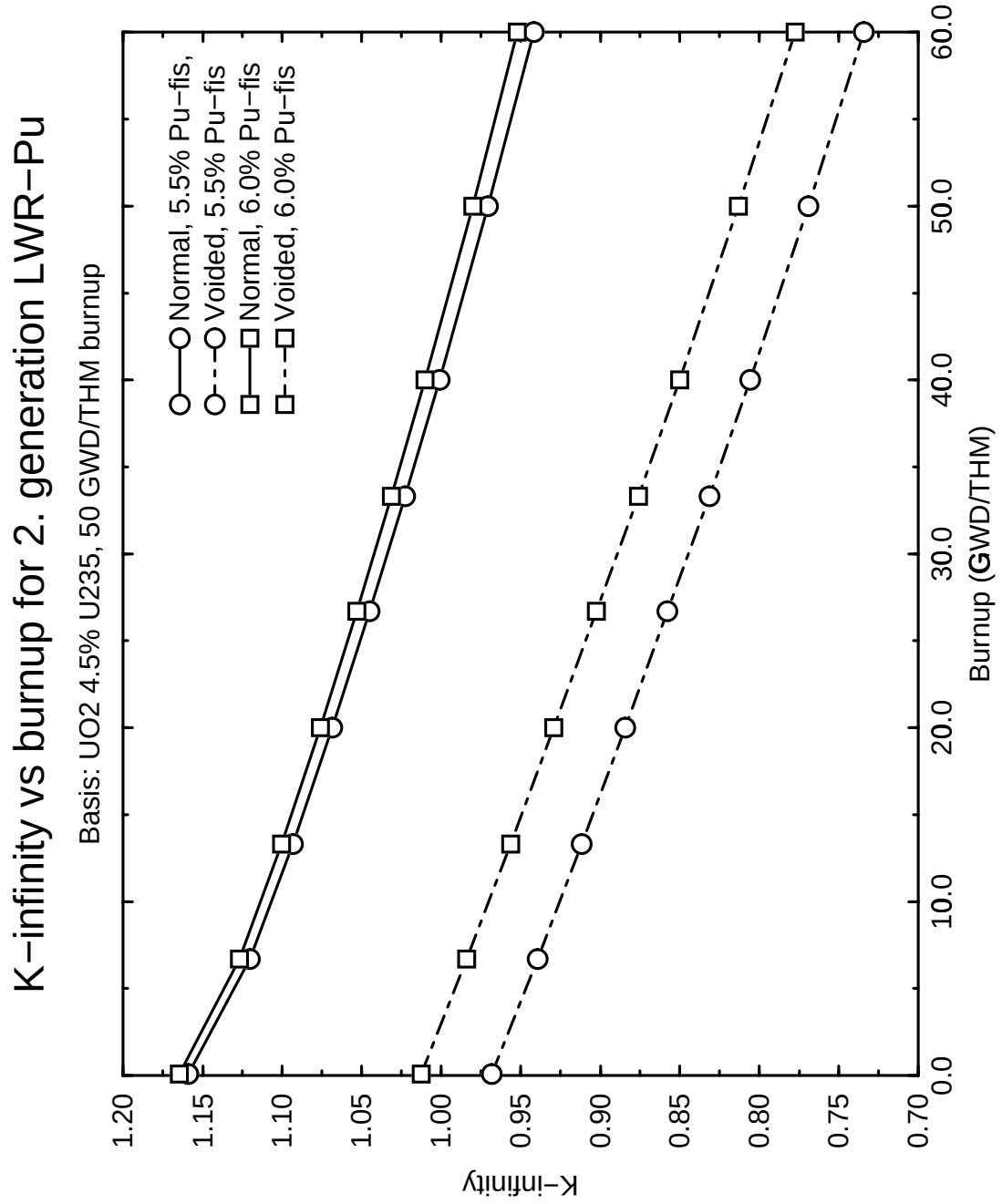

Figure 18: K_∞ void effect for 2. generation MOX, 50 GWD/THM discharge burnup.

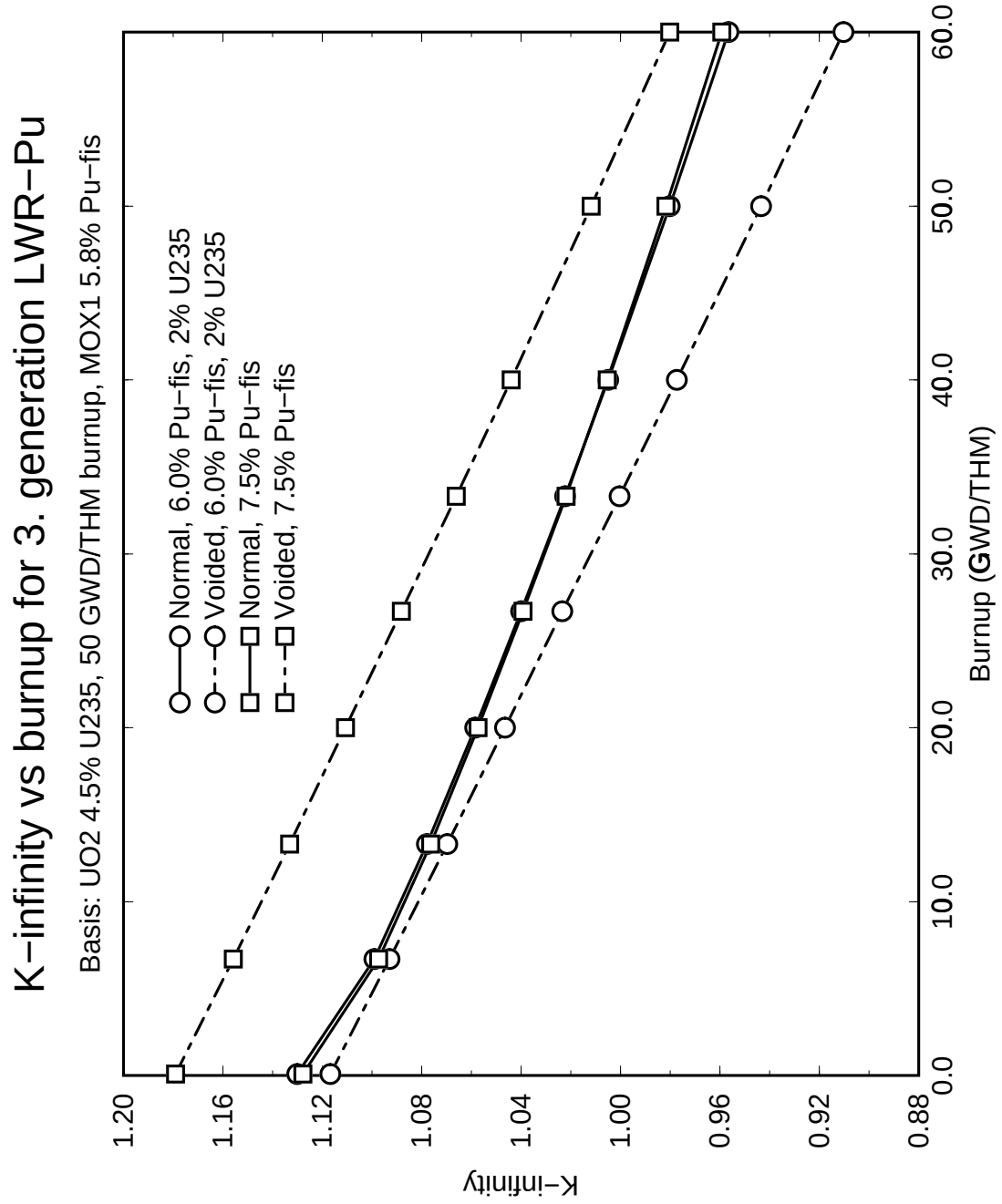

Figure 19: K_∞ void effect for 3. generation MOX, 50 GWD/THM discharge burnup.

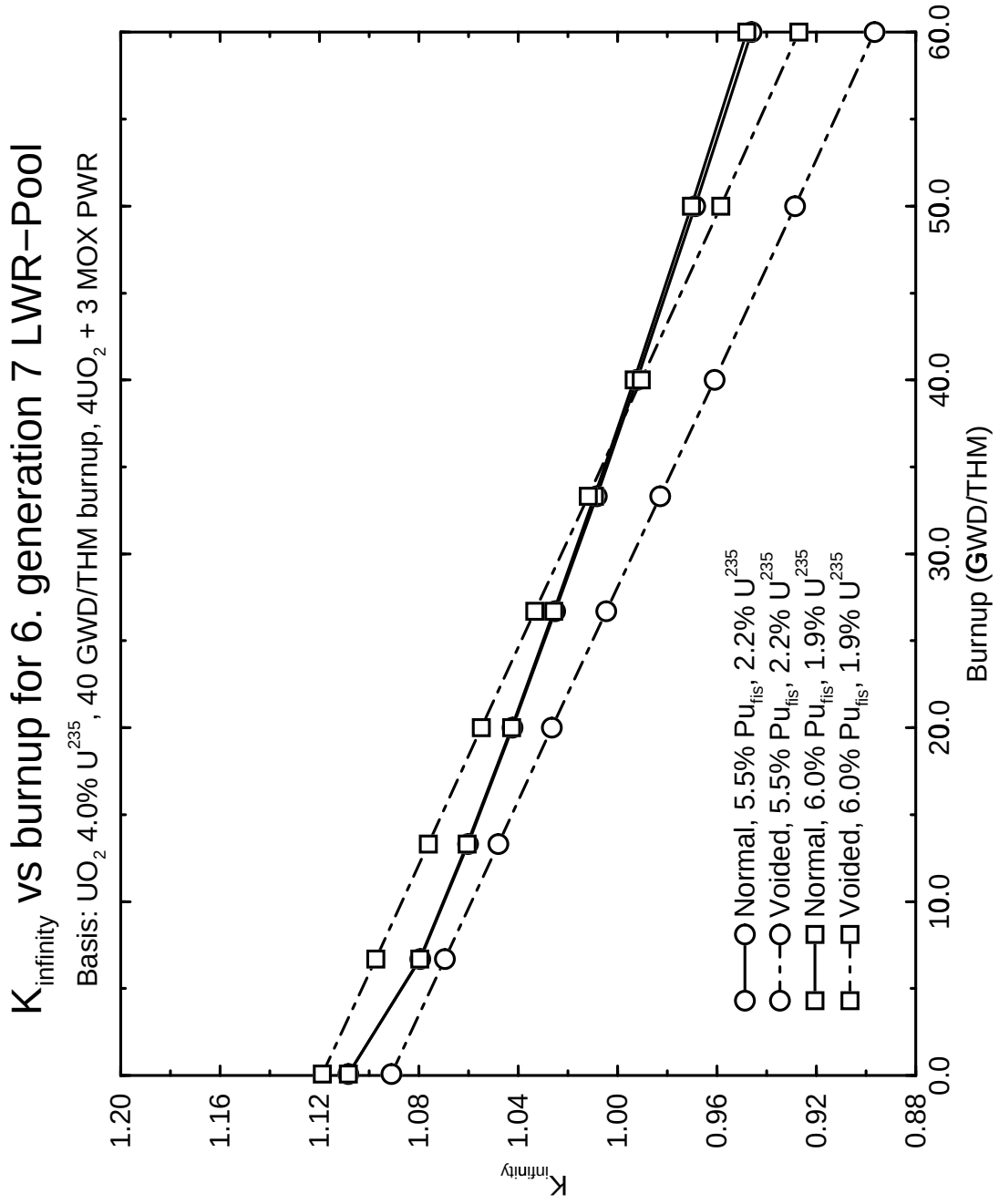

Figure 20: K_∞ Void effect for 6. generation MOX, 40 GWD/THM burnup.

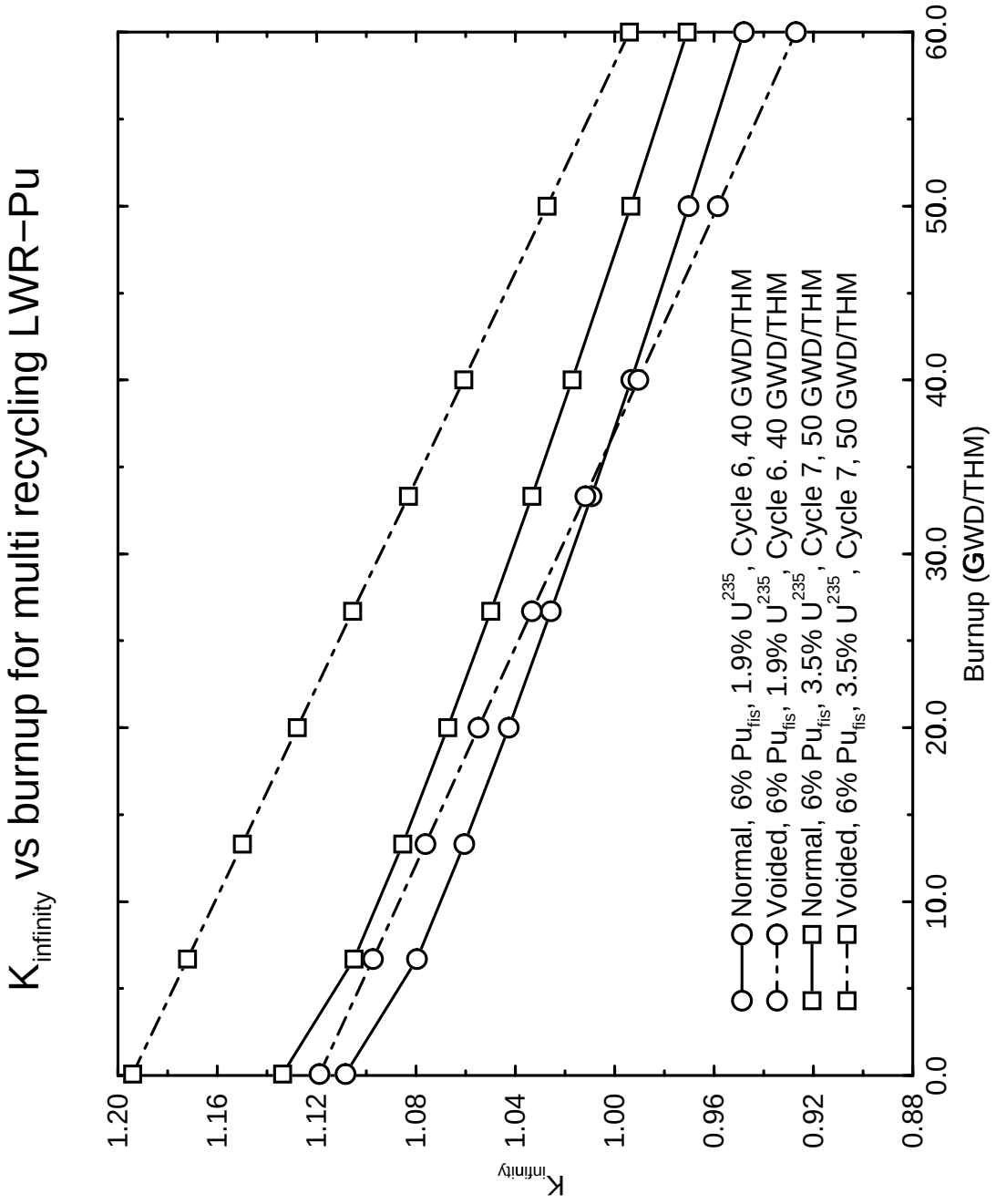

Figure 21: K_∞ Void effect for plutonium multi-recycling in PWRs.

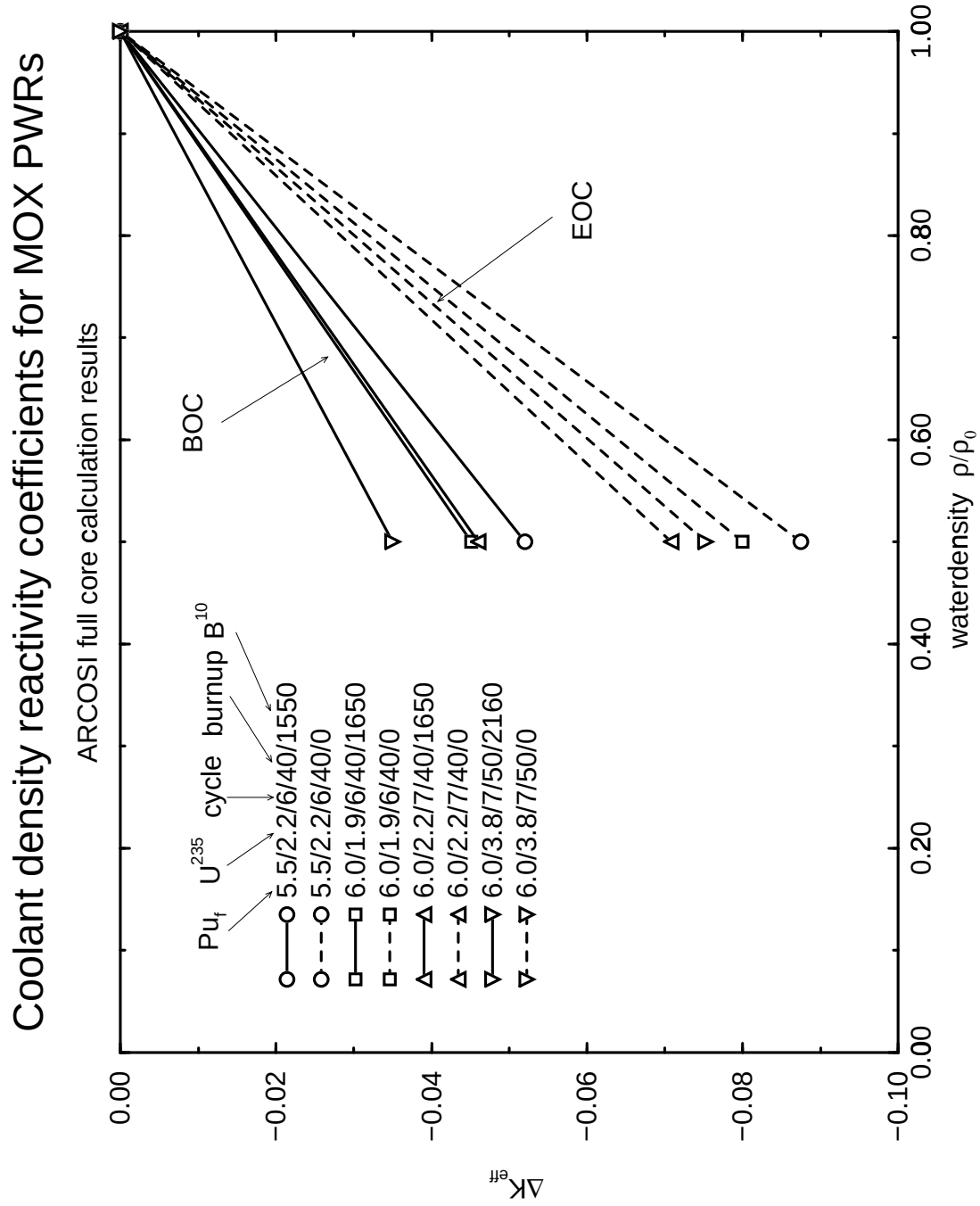

Figure 22: Coolant density reactivity coefficients for MOX PWRs.

7 Mass flow of transurania.

In this section the mass flows of the transurania isotopes are considered in more detail. First some basic properties of UOX and MOX lattices are summarized from earlier sections in order to remind the dependencies. Subsequently the transurania mass flows in PWR pools with plutonium multi-recycling according to the proposed scenario will be analyzed.

7.1 Transurania buildup in UOX and MOX lattices.

The transurania buildup in PWR lattices depends on lattice design and discharge burnup of the fuel. In reference [1] some systematic parameter variations for tight lattice light water reactors (APWR) are presented. The relevant parameter for transurania buildup in this study is the conversion ratio from fertile to fissile fuel isotopes. The conversion ratio generally increases for tighter lattices. This means that a wider reactor lattice with more moderator will lead to a reduction of the transurania buildup. In the present study no variation of the lattice pitch (distance between the centers of neighbouring fuel rods) has been performed. This is intended in later investigations.

The discussion of the tables 2 to 4 and of the figures 2 to 6 in section 3.3 give information about the buildup of transurania in UOX lattices of modern PWRs.

In the MOX lattices counterbalancing effects take place. On the one hand the fissile plutonium isotopes in the fuel are destroyed by fissions, on the other hand neutron captures in U^{238} and in the plutonium isotopes lead to the production of new plutonium isotopes. The tables 9, 13, 16 and 17 show net disappearance of plutonium for several fuel and burnup conditions. The shift of the plutonium isotopic distribution to the higher isotopes clearly may be observed in the tables 8, 12 and 15 and in the figures 4 to 6.

7.2 Transurania buildup in PWR pools.

In section 5.3 investigations for pools of PWRs with UOX and MOX cores have been performed. The transurania buildup per UOX and MOX reactor in these pools is summarized in table 18 for 3 target burnups and for up to 8 plutonium recyclings. The most significant effect is the considerable reduction of plutonium and the associated significant increase of the Am^{243} buildup in the MOX cores by more than a factor of 10.

The influence of plutonium multi-recycling on the buildup of transurania is shown in the figures 23 to 28. For the comparisons a normalization to GWe installed reactor power is chosen. This procedure takes into account that the pools have different number of reactors for the different target burnups. However, the different load factors for the fixed periods between reloadings are not considered when normalizing to generated electric power. In figure 23 the plutonium buildup is shown for a target burnup of 40 GWD/THM. The influence of the MOX cores clearly may be observed. In the case of 3 MOX cores contributing to the total

Reac-tor	Transurania in kg/TIHM								
	33 GWD/THM			40 GWD/THM			50 GWD/THM		
	Pu	Np^{237}	Am^{243}	Pu	Np^{237}	Am^{243}	Pu	Np^{237}	Am^{243}
U	+9.6	0.425	0.100	+10.6	0.543	0.126	+11.9	0.712	0.210
M1	-10.4	0.248	1.086	-14.7	0.287	1.405	-21.8	0.344	2.119
M2	-11.9	0.247	1.413	-16.2	0.286	1.776	-21.4	0.414	2.437
M3	-13.2	0.246	1.704	-17.6	0.285	2.101	-21.2	0.450	2.666
M4	-14.3	0.243	1.949	-19.4	0.281	2.495	-21.2	0.481	2.921
M5	-15.7	0.240	2.258	-18.7	0.313	2.641	-20.9	0.508	3.100
M6	-16.2	0.247	2.471	-18.4	0.350	2.891	-20.9	0.522	3.253
M7	-16.4	0.252	2.599	-18.5	0.354	2.989	-21.0	0.532	3.388
M8							-21.0	0.543	3.579

Identifications U,M1,M2..M7 from table 8.

Table 18: Transurania masses for pools of PWRs with different target burnups.

number of 7 PWRs, a net plutonium incineration takes place. After the 8 cycles a stabilisation of the plutonium inventory is found at a level of about half the value to be reached without recycling until that time. Figure 24 shows the same curves for 3 target burnups, 33, 40 and 50 GWD/THM. We can observe, that an increase of the target burnup leads to a small decrease of plutonium buildup if no recycling is applied (in-situ incineration of plutonium). In the recycling cases the tendencies for all target burnups are the same.

The figures 25 and 26 show the isotopic compositions of the plutonium depending on the number of recyclings. Both the unloaded and the loaded plutonium show a tendency to a steady state composition. The changes after about 8 cycles are fairly small.

The figures 27 and 28 show the buildup of Np^{237} and Am^{243} for the same cases as in figure 24. The Np^{237} buildup in figure 27 increases with increase of the target burnup, whereas the multi-recycling leads to a decrease. The influences only are moderate. The Am^{243} buildup in figure 28 shows a significant increase if without recycling the target burnup is increased from 40 to 50 GWD/THM. Much more pronounced is the increase due to the multi-recycling. In this case the buildups of Np^{237} and Am^{243} reach the same order of magnitude after about a century.

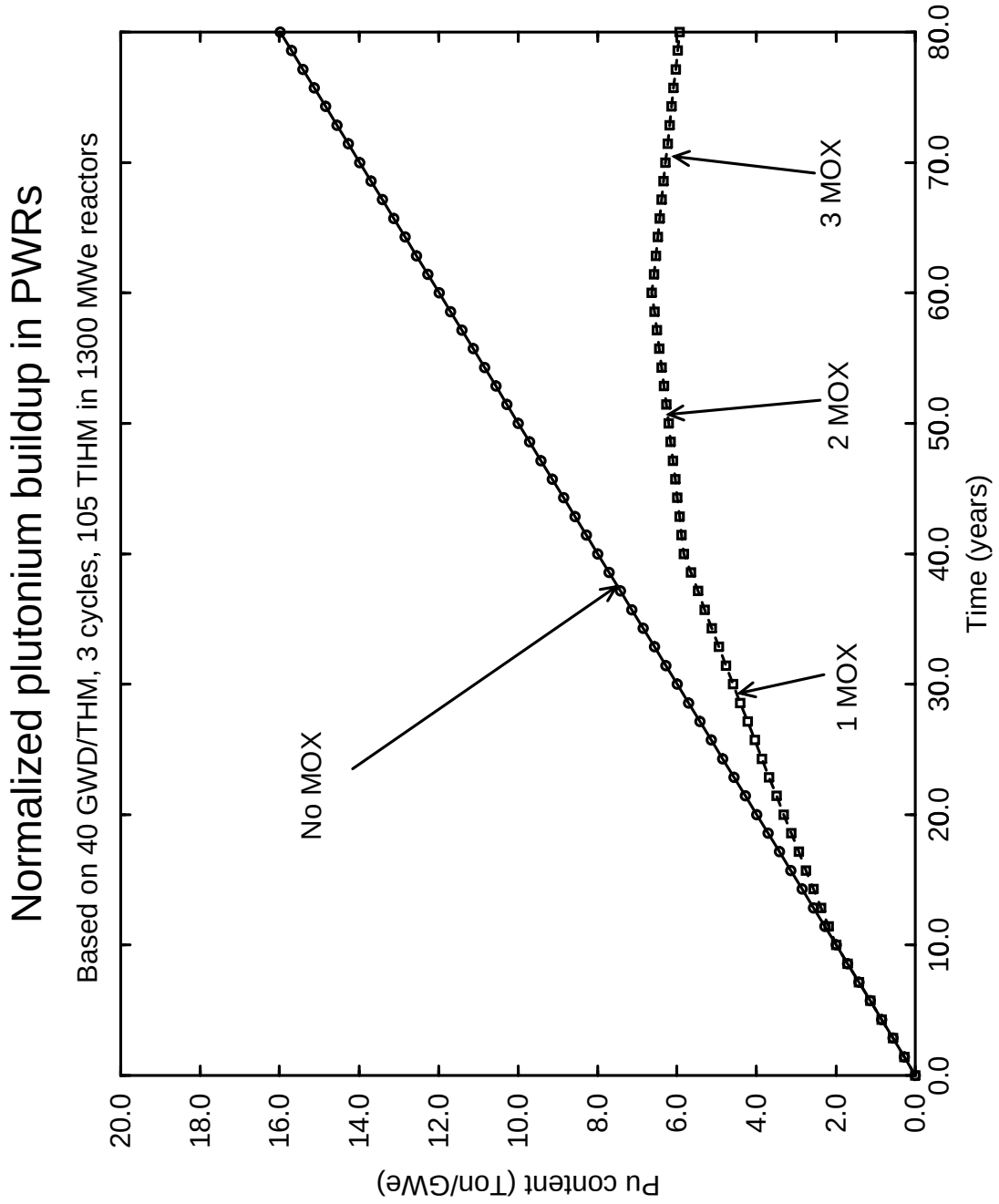

Figure 23: Plutonium buildup per GWe at 40 GWD/THM burnup in MOX PWRs.

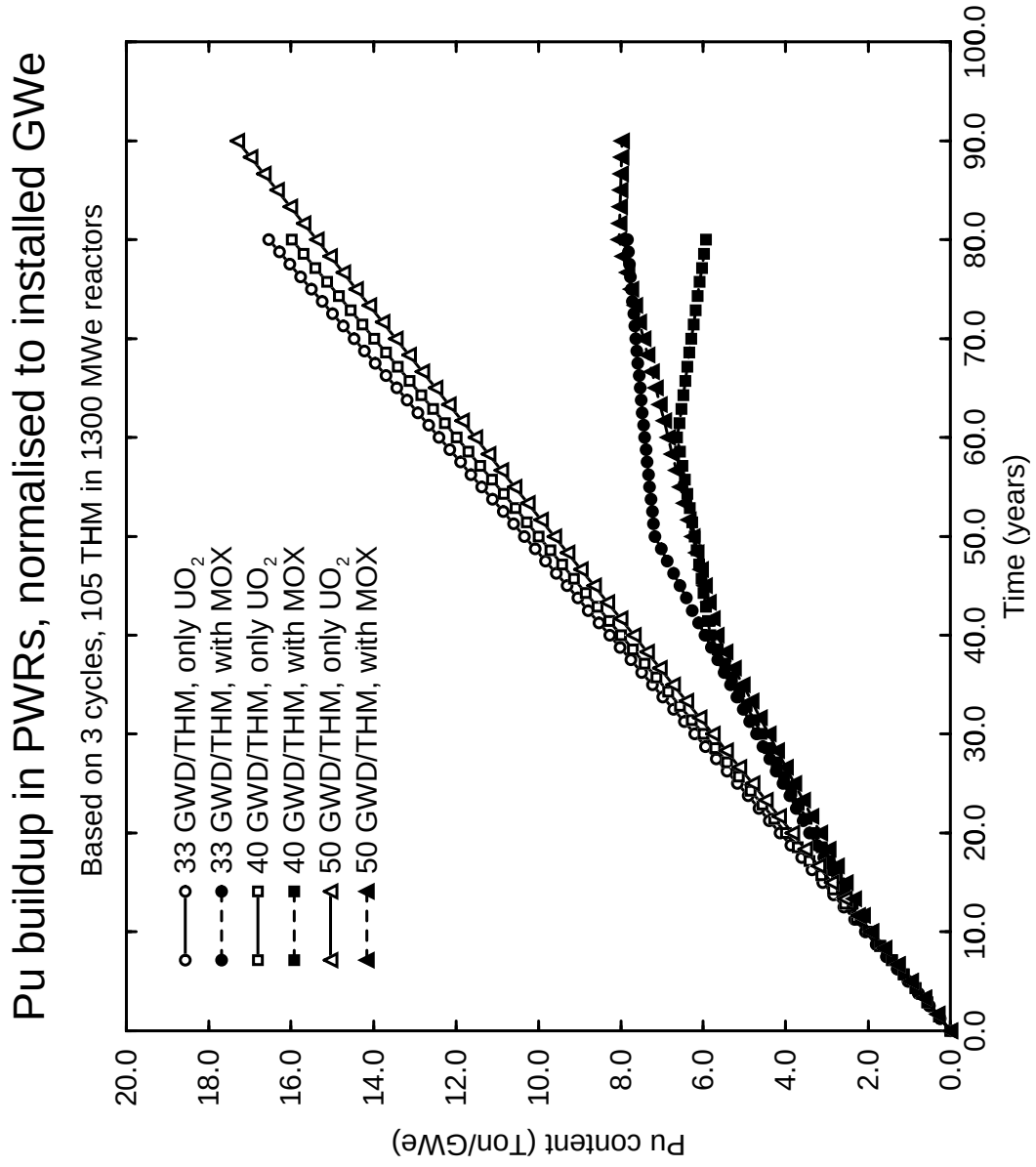

Figure 24: Plutonium buildup in PWRs for 33, 40 and 50 GWD/THM.

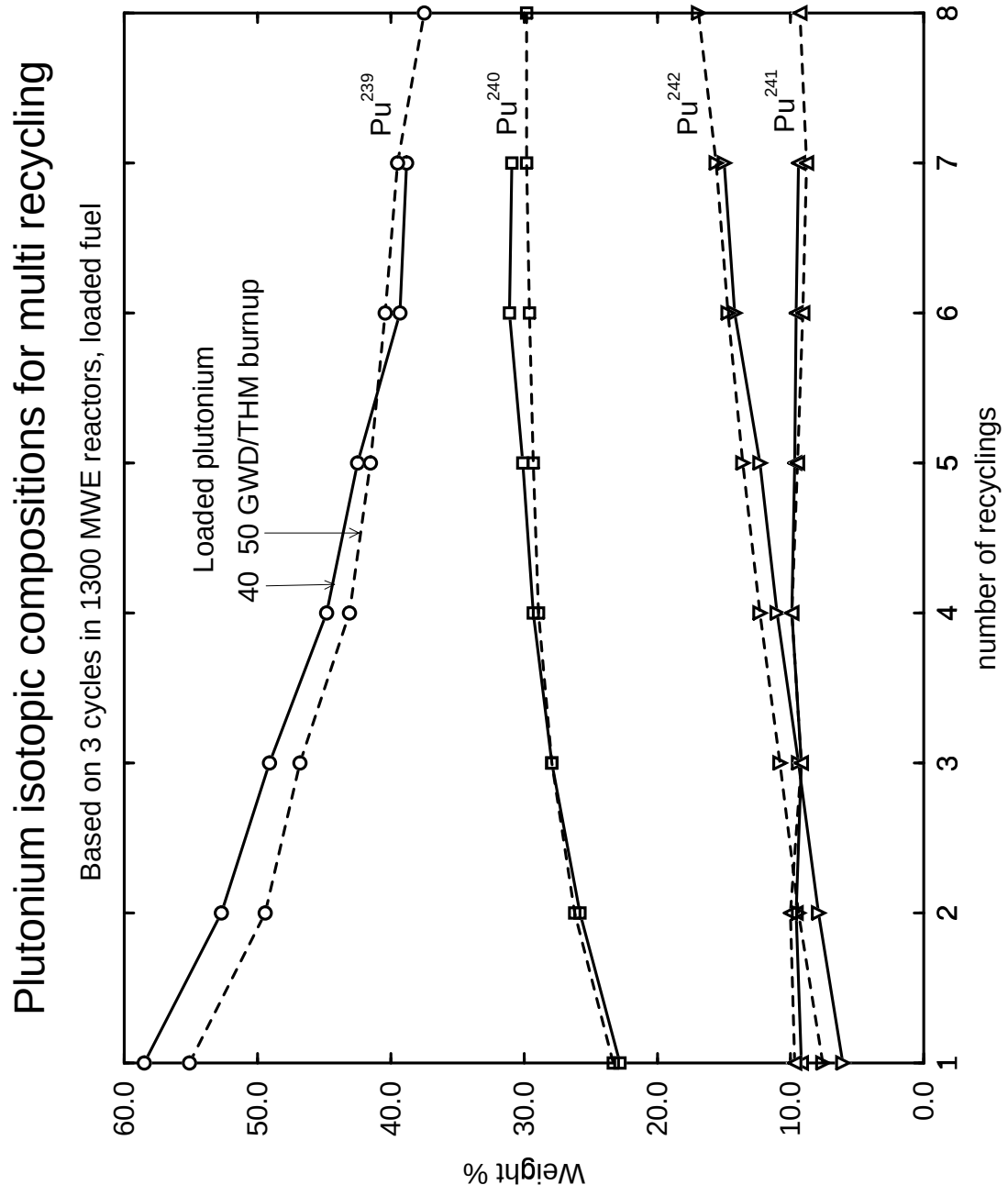

Figure 25: Changes of the composition of the loaded plutonium for multi-recycling in PWRs.

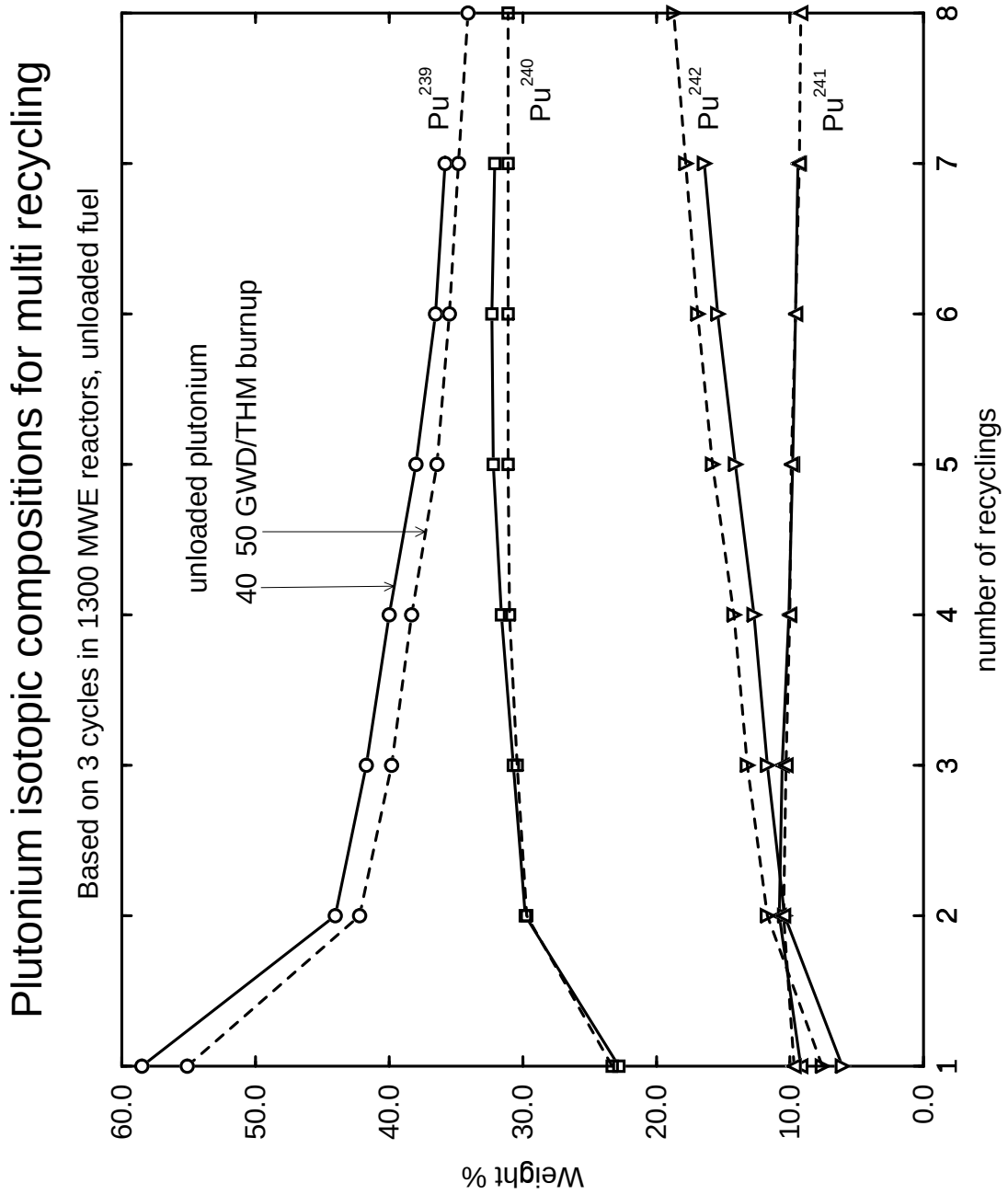

Figure 26: Changes of the composition of the unloaded plutonium for multi-recycling in PWRs.

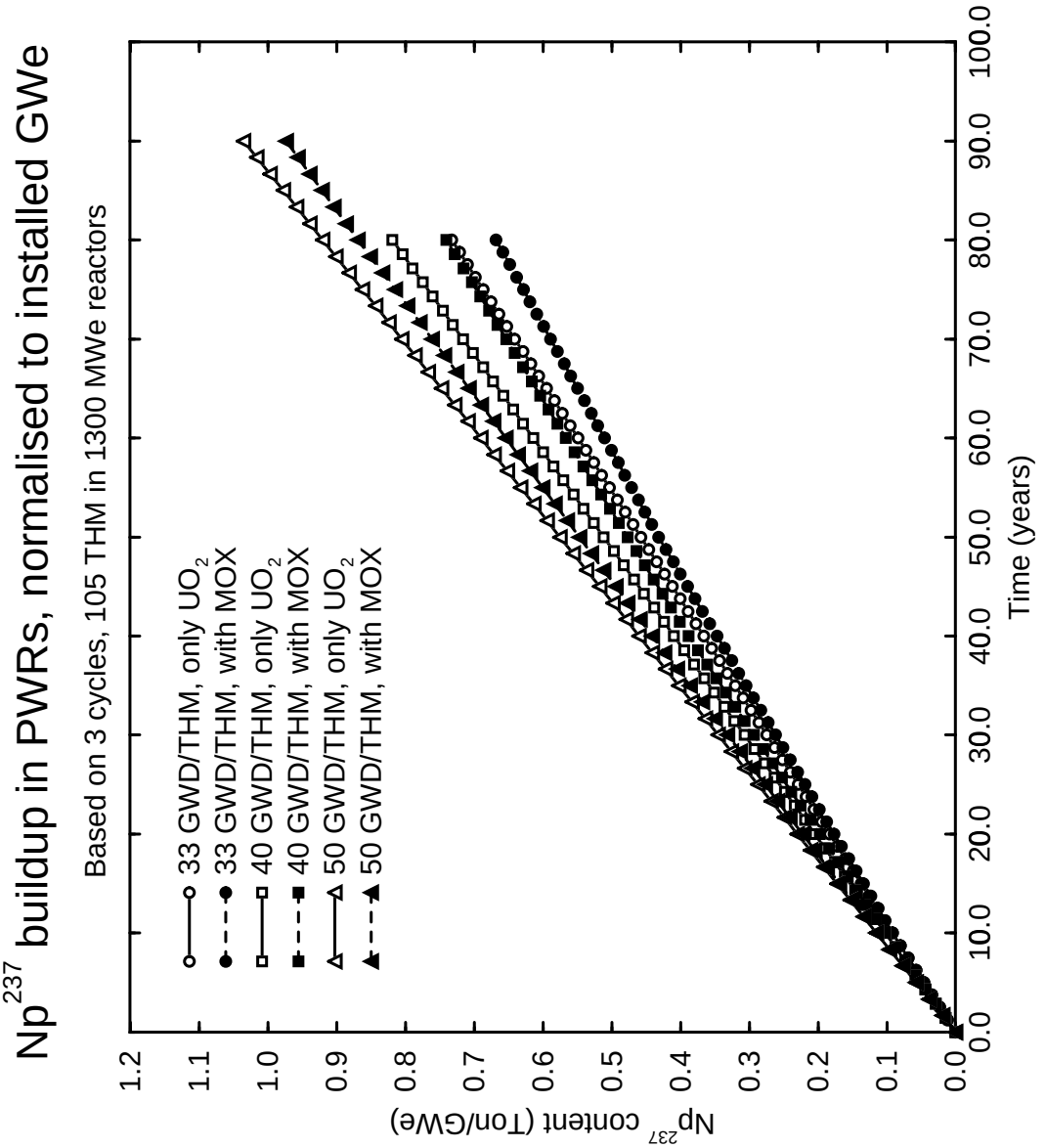

Figure 27: Np^{237} buildup per GWe in MOX PWRs.

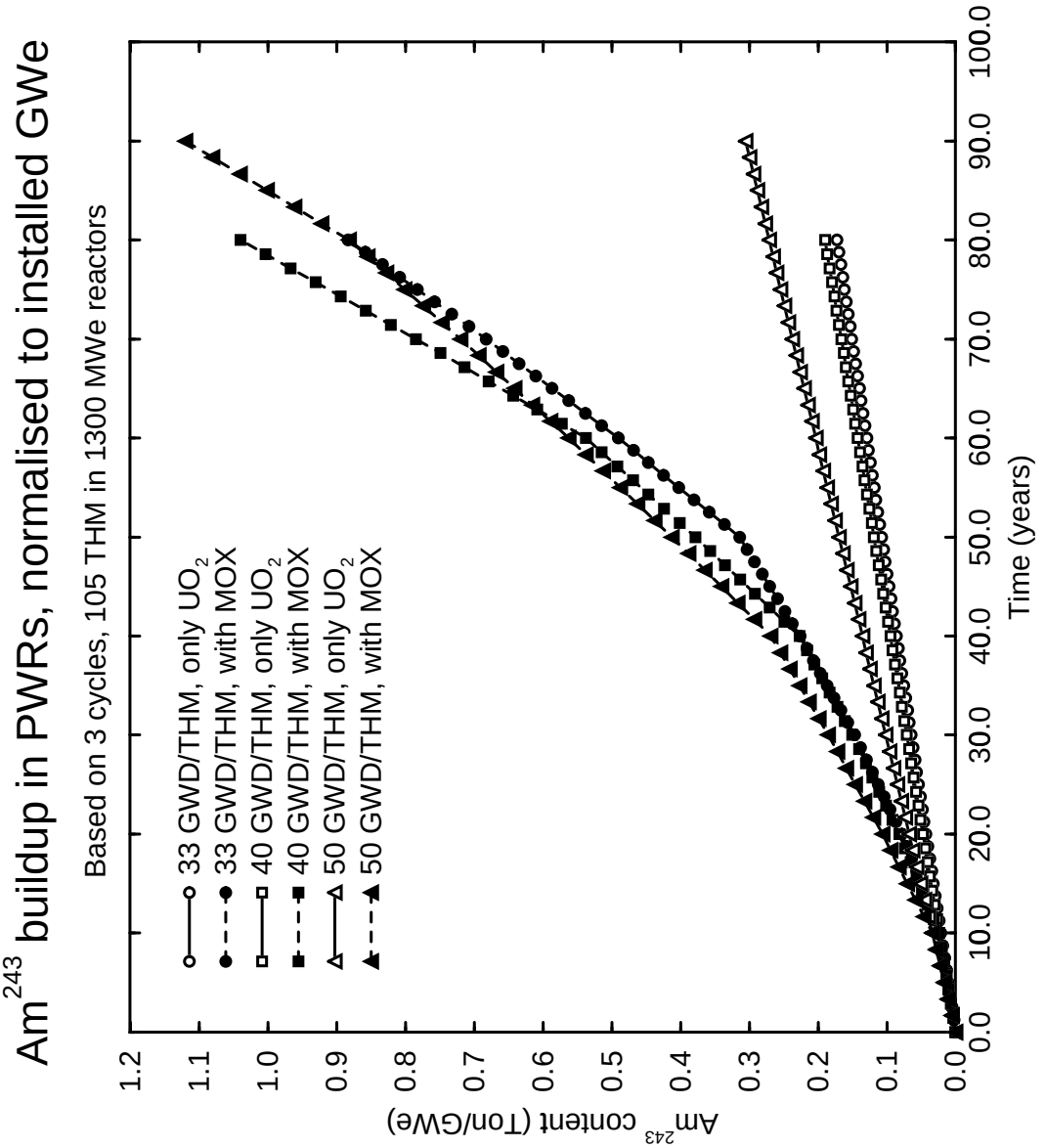

Figure 28: *Am*^{243} buildup per GWe in MOX PWRs.

8 The use of recycled uranium.

The results of section 7.2 indicate, that plutonium multi-recycling in PWRs may lead to a drastic decrease of the long term plutonium buildup. Prerequisite for such a scenario is the establishment of a closed fuel cycle with sufficient reprocessing and associated MOX refrabication capacity. However, during the reprocessing of spent nuclear fuel not only transurania but also large amounts of reprocessed uranium (RU) become available. The amount of RU in the spent fuel from a modern UOX PWR core is about 950 kg RU/THIM. Until now this RU usually goes into intermediate storage. The composition of the RU strongly depends on the starting conditions and on the irradiation behaviour. In most cases in UOX fuel assemblies from PWRs the U^{235} is burned out below about 1%. The (n,γ) reactions in U^{235} have lead to a significant buildup of U^{236}. Further the $(n,2n)$ and $(n,3n)$ reactions in the main heavy isotopes have produced small amounts of U^{232}, U^{233} and U^{234}. The ratio between U^{236} and U^{235} depends on the starting U^{235} enrichment in the fuel and on the discharge burnup and usually lies between $\approx$0.50 and $\approx$0.75. Similar phenomena occur during the irradiation of MOX fuel assemblies.

The main problem for the use of RU in PWRs is the buildup of the absorber isotope U^{234}. Moreover, the small U^{232} partition may cause problems due to the hard γ-radiation of its successor isotope Tl^{208}. The reuse of reprocessed uranium in PWR fuel assemblies may be realized by two alternative methods:

1. Enrichment of the RU to the required level for use in a PWR. In this procedure also U^{232} and U^{234} are enriched. Recently some activities are ongoing to make available enrichment capabilities for RU [21].

2. Blending high enriched uranium with the RU to obtain the required fuel enrichment. With this solution additional amounts of depleted uranium are produced.

Both methods have certain advantages and disadvantages. Enrichment of the RU needs transport from the reprocessing unit to the enrichment unit. This transport route may be kept short if both units are in the same nuclear fuel cycle park. Blending of high enriched uranium with the RU needs higher enriched uranium with high separation costs and possible problems regarding non-proliferation aspects. In the next sections estimates for the required enrichments and for the amounts of the uranium to be mixed with the RU are discussed. First preliminary costs estimates are also given.

8.1 Estimates for the uranium to be mixed with RU.

The use of fresh uranium requires U^{235} enrichments varying from $\approx$3.2% for a target burnup of 33 GWD/THM to $\approx$4.5% for a target burnup of 50 GWD/THM. The reprocessed uranium RU may be converted to useable fuel for a PWR by mixing it with a rather small

fraction of fairly high enriched fresh uranium. If we neglect the reprocessing losses as a first approximation, the following expression is valid:

$$F \cdot f_F = O \cdot f_O + B \cdot f_p \tag{18}$$

with

F	total load quantity
O	quantity of reprocessed uranium (RU) from the preceding reactor cycle
B	quantity of fresh higher enriched uranium
f_F	enrichment in the reloading fuel
f_O	enrichment RU
f_p	enrichment fresh uranium

Further we have

$$F = O + B \tag{19}$$

From (18) and (19) follows

$$f_p = f_F + \frac{O \cdot (f_F - f_O)}{B} \tag{20}$$

In table 19 some results of inventory calculations for UOX fuel assemblies in PWRs are summarized for 3 burnup stages. Tabulated are the BOC and EOC compositions of the uranium, the EOC ratio U^{236}/U^{235} and the characteristics of the needed fresh uranium. For these calculations the assumption is made, that the enrichment for the reloading cores is constant. The results for the succeeding MOX cores show, that the buildup of U^{234} leads to the need for a small enrichment increase.

In table 20 results for selected cases from the investigations for pools of PWRs in section 5.3 are summarized. The selected reactor cycles 7 and 9 are near to equilibrium cycles. For the calculations it is assumed that the reprocessed uranium from these selected cycles is reused for the first time, i.e. not from the beginning of the plutonium recycling. In addition to the parameters of table 19, information about the number of the plutonium recyclings and about the reactivity impact of the U^{234} and U^{236} isotopes is given. ΔK_{RU} gives the differences in K_∞ if instead of fresh, reprocessed uranium is used. The last row of table 20 shows that for the 9. plutonium recycling for a target burnup of 50 GWD/THM, the U^{235} enrichment of the fresh uranium must be increased by about 0.2% to obtain the same reactivity at the end of cycle if RU is used.

8.2 Cost estimates for use of reprocessed uranium.

The use of reprocessed uranium (RU) may lead to the need for fairly high enriched uranium to be mixed with the RU in order to get sufficient reactivity. The separation costs for the uranium enrichment increases more than proportial with the required enrichment. On the basis of the following data, cost estimates have been made in reference [22].

U_{nat}	0.71% U^{235}
U_{tail}	0.25% U^{235}
Uranium separation work (USW)	100 \$/kg
Natural uranium + conversion	30 \$/kg

From these specifications, in table 21 crude estimates are made for the costs of **fresh enriched uranium** and of **RU blended by high enriched fresh uranium**. The numbers are related to 1 ton of uranium. These estimates only concern the uranium material costs without considering possible consequences for the fuel fabrication. Cost estimates for the enrichment of RU have not been performed. We may observe, that for the UOX fuel assemblies no significant differences occur. In the case of MOX fuel assemblies with enriched U^{235} the use of reprocessed uranium seems to be favourable.

Important advantages of the reuse of reprocessed uranium are the saving of uranium resources and the reduction of storage capacity for spent fuel.

Burnup (GWD/ THM)	BOC U^{235} (%)	EOC uranium vector (%)				Ratio U^{236}/U^{235}	Fresh ⋆ U^{235} per TIHM	
		234	235	236	238		(kg)	(%)
33	3.2	0.00	0.84	0.43	98.73	0.51	44.7	53.6
40	4.0	0.00	1.04	0.56	98.40	0.54	52.8	57.1
50	4.5	0.00	0.92	0.67	98.41	0.73	64.6	56.3

⋆ Results from formula (20) for f_F = constant.

Table 19: Selected results for UOX loadings in PWRs.

Burnup (GWD/ THM)	Cycl.	BOC U^{235} (%)	EOC uranium vector (%)				Ratio U^{236}/U^{235}	Fresh ⋆ U^{235} per TIHM		EOC ‡ ΔK_{RU}
			234	235	236	238		(kg)	(%)	
33	7	1.0	0.01	0.70	0.08	99.21	0.11	22.1	12.7	-0.0013
40	7	2.0	0.01	1.33	0.19	98.47	0.14	27.8	22.6	-0.0023
50	7	3.3	0.02	2.04	0.35	97.61	0.17	36.5	32.5	-0.0040
50	9	3.8	0.02	2.39	0.39	97.20	0.16	37.0	35.7	-0.0038
50	9	4.0 †	0.03	2.54	0.76	96.66	0.30	37.6	36.6	-0.0007 +

⋆ Results from formula (20) for f_F = constant.
‡ $\Delta K_{RU} = K_{\infty,noU6} - K_{\infty,withU6}$
† RU loading with 0.69% U^{236} and 0.02% U^{234}
+ Related to 3.8% U^{235} enrichment without U^{236}

Table 20: Selected results for MOX loadings in PWRs.

FA-type	Burnup (GWD/THM)	Fresh uranium				RU			
		% U5	kg	$/kg	$	% U5 ⋆	kg ⋆	$/kg	$
UOX	33	3.2	1000	610	610.000	53.6	44.7	15.490	690.000
UOX	40	4.0	1000	830	830.000	57.1	52.8	16.550	870.000
UOX	50	4.5	1000	960	960.000	56.3	64.6	16.310	1.054.000
MOX	33	1.0	1000	80	80.000	12.7	22.1	3.300	73.000
MOX	40	2.0	1000	305	305.000	22.6	27.8	6.200	172.000
MOX	40	3.3	1000	640	640.000	32.5	36.5	9.200	336.000
MOX	50	3.8	1000	770	770.000	35.7	37.0	10.100	374.000
MOX	50	4.0	1000	830	830.000	36.6	37.6	10.400	391.000

⋆ Results from formula (20) for f_F = constant.

Table 21: Comparison of the fuel costs for fresh and reprocessed uranium for PWRs.

9 Summary.

The back end of the nuclear fuel cycle is of large importance for the near future. The spent fuel from nuclear power plants contains large amounts of heavy isotopes and fission products with partially very long decay times. The atomic act in Germany prescribed for a long period of time the recycling of the spent reactor fuel for the use for further energy production. However, a recent amendment also allows the direct disposal of the spent fuel. The objective of the present investigations is to gain knowledge for the judgement of possible options for the back end of the nuclear fuel cycle. The main emphasis is directed to the the pressurized light water cooled reactors (PWRs), being currently the most utilized reactor type.

In section 2 the characteristics of important heavy isotopes are discussed. For long term investigations mainly most of the plutonium isotopes and Np^{237}, Am^{241} and Am^{243} have to be considered in more detail. For very long irradiated fuel also the curium isotopes $Cm^{245\ 246\ 247}$ may become of interest.

In section 3 basic investigations for fuel lattices of modern PWRs are described. The applied calculational procedures are discussed in some detail. Parametric investigations are carried out to analyze the transurania buildup in UOX lattices in dependence of the fuel enrichment and of the discharge burnup. Generally, an increase of the discharge burnup in PWRs, due to the in-situ burning of plutonium, leads to a decrease of the specific plutonium production and to an increase of the neptunium and the americium buildup. The fissile content of the plutonium decreases with increasing burnup. An important question for the use of plutonium in PWRs is the equivalency of fuel assemblies with uranium and mixed uranium/plutonium oxide in the same PWR core, because this is the actual applied method for the utilization of the plutonium coming from nuclear fuel reprocessing plants. On the basis of experiences with MOX fuel in German and French PWRs, a relation for the end of cycle values of K_∞ in both fuel types could be specified. The new relation leads to lower values for the required plutonium enrichment of the MOX fuel, compared to the results in earlier studies.

In section 4 exploratory whole core calculations for full MOX PWRs are presented. The calculational procedures for these investigations are discussed in some detail. They have been developed for earlier studies for advanced pressurized water reactors [1]. The primary aim of the present whole core calculations is to investigate the capabilities for plutonium multi-recycling in PWRs. The results show, that plutonium multi-recycling in PWRs is feasable for a limited number of recyclings if depleted or natural uranium is to be used for the MOX fuel and safety related parameters have to be maintained. The allowed number of recyclings depends mainly on the discharge burnups of the fuel assemblies, on the mixing strategies for the recycled plutonium and on the quality of the uranium. The detoriation of the coolant density reactivity coefficients limits the plutonium fissile fraction of the MOX fuel to $\approx$6%.

In section 5 long term considerations for plutonium recycling in PWRs are presented. These investigations are based on a scenario with pools of PWRs, consisting of only UOX cores at the beginning. As soon as enough plutonium is produced in this pool, UOX cores are replaced by full MOX cores. For the ex-core times, 7 years of cooling and reprocessing

time and 3 years of fabrication time are chosen. The investigations have been performed for discharge burnups of 33, 40 and 50 GWD/THM. The plutonium for the next cycle is obtained by mixing all available plutonium from the UOX and the MOX cores. The fissile fraction of the plutonium is limited to ≈6%; if neccessary enriched U^{235} is used to meet criticality conditions. This scenario for plutonium multi-recycling leads to a near equilibrium plutonium composition with constant inventory after about 100 years. At this time the amount of plutonium is about half the value, compared to a scenario without plutonium recycling.

In section 6 first investigations for safety related parameters are discussed. The most relevant safety parameters are the fuel temperature reactivity effect (Doppler effect) and the coolant density reactivity effect. In reference [1] it was found, that the Doppler effect is not very sensitive to the change from UOX to MOX fuel. Therefore no special investigations were performed for this study. For the coolant density reactivity coefficients a first indication may be obtained by calculations for voided lattices. If such calculations give sufficiently negative reactivity effects, as e.g. for UOX lattices, no further investigations are required. If the fissile plutonium content of the MOX fuel exceeds ≈6%, the lattice voiding effects change from less negative to positive. In such cases more accurate whole core calculations are necessary. For selected cases from the preceding whole core calculations, first coolant density reactivity effects have been determined. With the help of special data libraries with reduced coolant densities, the reactivity changes at the begin and the end of the reactor cycle have been calculated for half the nominal coolant density. The results of these investigations indicate, that no problems with coolant density reactivities occur for the reactors of the pool scenario, described before.

In section 7 the mass flows of the transurania have been analyzed in more detail. For the buildup of transurania in a scenario without plutonium recycling the influence of the discharge burnup and of the U^{235} enrichment have been considered. Higher discharge burnups lead to more in-situ incineration of plutonium with the consequences of less specific amounts of plutonium of worse quality with a smaller fission fraction and of more buildup of neptunium and especially americium. Higher U^{235} enrichments act against these trends. The influence of plutonium recycling on the mass flows of the transurania has been studied for the pool scenarios, mentioned before. As stated above, after about 80 years a nearly constant plutonium inventory may be reached at about half the level without recycling at that time. This means that large savings of plutonium buildup may be obtained by plutonium recycling in PWRs. The buildup of neptunium is not sensitive to plutonium recycling. However, the buildup of americium reaches the same order of magnitude as the neptunium if plutonium recycling is applied. Otherwise the amounts of americium are significantly smaller than the amounts of neptunium.

In the last section 8 some aspects of the reuse of recycled uranium from the spent fuel are discussed. The buildup of the absorber isotope U^{234} is disadvantageous for the use in PWRs. First estimates indicate the need of an increase of the U^{235} enrichment of ≈0.2% if uranium after 1 reprocessing is used. For the enrichment of reprocessed uranium, 2 alternative methods have been analyzed: treating the reprocessed uranium in an enrichment facility or mixing the reprocessed uranium with highly enriched fresh uranium. Preliminary costs estimates in-

dicate, that both methods are comparable for the reprocessing of UOX fuel assemblies. In the case of MOX fuel assemblies, the blending of the reprocessed uranium with highly enriched fresh uranium seems favourable.

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