# Investigations on fuel utilization and transaranium buildup in PWRs

**C.H.M. Broeders**

*Forschungszentrum Karlsruhe, Postfach 3640*
*D76021 Karlsruhe, Germany*

**Abstract.** Due to the large amounts of synthetic non-stable heavy isotopes and fission products, the back end of the nuclear fuel cycle is a serious problem for the near future. The buildup of transurania in pressurized water reactors (PWRs), being the most utilized reactor type at present, is investigated without and with plutonium recycling in PWRs. The applied calculational procedures have been developed previously for the investigation of tight lattice light water reactors and are validated carefully for reactor systems with fast, thermal and epithermal neutron spectra. If no plutonium recycling is applied, the transurania buildup decreases slightly and the fractions of neptunium and americium increase significantly if the fuel discharge burnup is increased. A plutonium multirecycling scenario is presented with pools of full uranium oxide (UOX) and full mixed uranium and plutonium oxide (MOX) cores that leads to a drastic decrease of the plutonium buildup. After 60 to 80 years, the plutonium inventory in the pool stabilizes at a level of about 50% of the inventory without recycling at that time. Possible safety problems with coolant density reactivity coefficients are avoided by the limitation of the plutonium content and by the use of enriched uranium in the MOX fuel if required for reactivity reasons.

## INTRODUCTION

One of the most severe problems for the peaceful use of nuclear energy is the back end of the nuclear fuel cycle. Neutron absorptions in the heaviest natural element U cause the buildup of non-stable transurania elements, mainly Np, Pu, Am and Cm. Some of these transurania isotopes have good fission properties and are well suited for reuse in nuclear reactors. Furthermore, the fission of the heavy elements leads to the generation of several long-lived fission products, e.g. Tc, Cs and I. Because of their toxity and radiation hazards, these new synthetic materials are serious sources of potential danger for mankind. Besides the outcomes from nuclear reactors for civil utilization, the military programs of several countries in the world also have produced quite large amounts of transurania and fission products. One of the objectives of the R/D program at the Forschungszentrum Karlsruhe (FZK) is to maintain the capabilities for the assessment of the possible options to reduce the impact of the back end of the nuclear fuel cycle, see e.g. Ref. [1]. In the present contribution the buildup of transurania in pressurized water reactors (PWR), being nowadays the most widely used reactor type, is investigated. More information may be found in Ref. [2].

## APPLIED CALCULATIONAL PROCEDURES

The applied calculational procedures are described in some detail in Ref. [3] and are collected in the code system KARBUS. They combine advantages of the standard methods for thermal and fast reactors and have been developed for the investigation of tight lattice light water reactors, including the neutronic calculation of basic fuel lattices, of subassemblies (SA) and of whole reactor cores. The fuel lattice calculations use first flight collision probability methods from the WIMS code [4]. The whole core calculations are performed for the equilibrium state of a PWR with hexagonal SA with the nodal code HEXNOD [5]. The SA cross section data for the present exploratory investigations were directly taken from lattice calculations, neglecting the SA structures and irregular water gaps. The reactor depletion calculations use the formalisms of the widely used ORIGEN code [6]. The applied 69 group libraries are mainly based on the latest KEDAK4 evaluations by FZK for the common reactor materials in coolant, structures and fuel and on JEF-2 data for about 80 fission products. These calculational procedures have been carefully validated for reactor systems with fast, thermal and epithermal neutron spectra, see e.g. Ref. [3]. Recently a comparison between KARBUS and the French standard calculational methods for PWRs showed good agreement in the results for a benchmark investigation for Pu multi-recycling in PWRs, see e.g. Ref. [7].

## BUILDUP OF TRANSURANIA IN PWRs WITHOUT FUEL RECYCLING

Modern PWRs contain about 80 tons of heavy metal fuel per gigawatt electrical power (GWe). Depending on the characteristics of the nuclear power station (discharge burnup, fuel management, power ratings) $\approx \frac{1}{3}$ of the core is unloaded every 1. to 1.5 years. In this section some estimates are given for the buildup of transurania in this unloaded fuel. The results are normalized to 1 ton of initial heavy metal (TIHM). For these investigations only lattice calculations for the basic cell of a modern PWR are performed. The discharge burnups have been varied from 33 gigawatt days per ton heavy metal (GWD/THM), being representative for the today burnups and 50 GWD/THM, being a near future target burnup from economical considerations. For these burnups in modern PWRs, reasonable enrichments for the $U^{235}$ vary between 3.2 and 4.5%. In table I the results of these parametric investigations are summarized. The buildup of transurania amounts between 10 and 13 kg/THIM. More than 90% of the transurania consists of Pu. The Np fraction amounts to 4..6%, the Am fraction to 1..2%. The weights of $Np^{237}$ and $Am^{243}$ increase strongly with burnup increase. The fissile fraction of the Pu decreases significantly with burnup increase.

## TRANSURANIA IN A POOL OF PWRs WITH PU MULTI-RECYCLING

The use of fuel assemblies with mixed U and Pu oxide (MOX) fuel is proven technology in PWRs, e.g. in France and in Germany. Until now mainly good quality Pu with high fissile fractions has been utilized for discharge burnups of ≈35 GWD/THM. The main result of the investigations described in Ref. [7] is, that after a small number of recyclings the composition of the Pu becomes so unfavourable, that the fissile enrichment of the Pu has to exceed 6..7% if about 50 GWD/THM discharge burnup should be reached with natural or depleted U in the MOX. With such Pu fractions in the MOX fuel it cannot be guaranteed, that the important safety parameter 'coolant density reactivity coefficient' (CDRC) remains satisfactory [3]. Starting from this experience, in Ref. [2] whole core calculations for pools of PWRs with full U oxide (UOX) and full MOX cores have been performed. To avoid problems with the CDRC, the fissile fraction of the Pu was limited to ≈6%. In order to get enough excess reactivity at reactor startup to obtain the required discharge burnups, $U^{235}$ enriched uranium is used if necessary. The main result of these investigations is that, independent of the discharge burnup, the ratio between the Pu production in an UOX core and the Pu incineration in a MOX core is nearly constant: ≈0.6. This means, that the combined operation of 5 UOX and 3 MOX cores leads to a nearly constant Pu inventory in the pool of 8 PWRs. The Pu composition in such a pool only changes very slowly; a near to equilibrium state may be reached after 60 to 80 years. The enrichment of the $U^{235}$ in the MOX with 6% fissile Pu strongly increases with increasing discharge burnup and varies from ≈1% at 33 GWD/THM to ≈4% at 50 GWD/THM.

## PWR TRANSURANIA MASSES WITHOUT AND WITH PU RECYCLING

In order to estimate the transurania masses, a number of calculations for pools of PWRs with UOX and MOX cores have been performed for different discharge burnups. The ex-core time of the fuel was chosen to be 10 years: 7 years cooling / reprocessing and 3 years fabrication time. Within these 10 years a distinct number of reactor core cycles has to be determined, mainly depending on the discharge burnup. The total number of reactors in a pool was fixed by the requirement, that after the first ex-core cycle of the fuel, one UOX core may be replaced by a full MOX core with the Pu produced. For the discharge burnups 33, 40 and 50 GWD/THM up to 9 recyclings have been studied by whole core calculations. The main results are presented in the figures 1 and 2. The produced transurania are normalized to 1 GWe installed. In figure 1 the buildup of Pu shows a significant decrease if multi-recycling is applied. The discontinuities in the curves with MOX indicate the replacement of a UOX core by a MOX core in the pool. After 60 to 80 years we may obtain a nearly constant Pu inventory at a level of about 50% of the inventory without recycling at that time. This means also, that comparable amounts of U may be saved for later use. In figure 2 the buildup of $Am^{243}$ shows a significant increase due to the multi-recycling of the Pu. In these cases the buildup of $Am^{243}$ is comparable with the buildup of $Np^{237}$, being not sensitive to the Pu recycling.

## References

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Table I: Transurania buildup and Pu compositions in the unit cell of a modern PWR for different discharge burnups and U enrichments

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

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

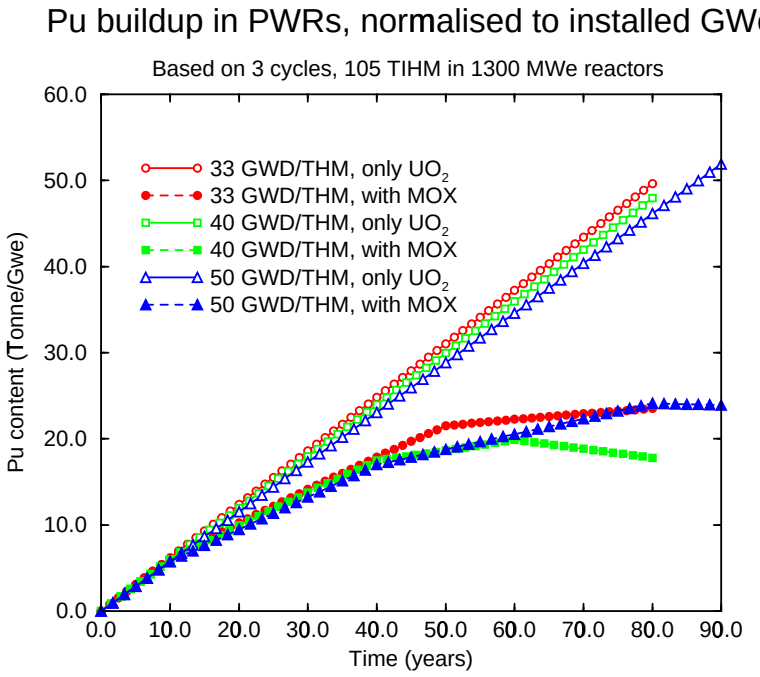


Figure 1: Plutonium buildup in PWRs for burnups of 33, 40 and 50 GWD/THM

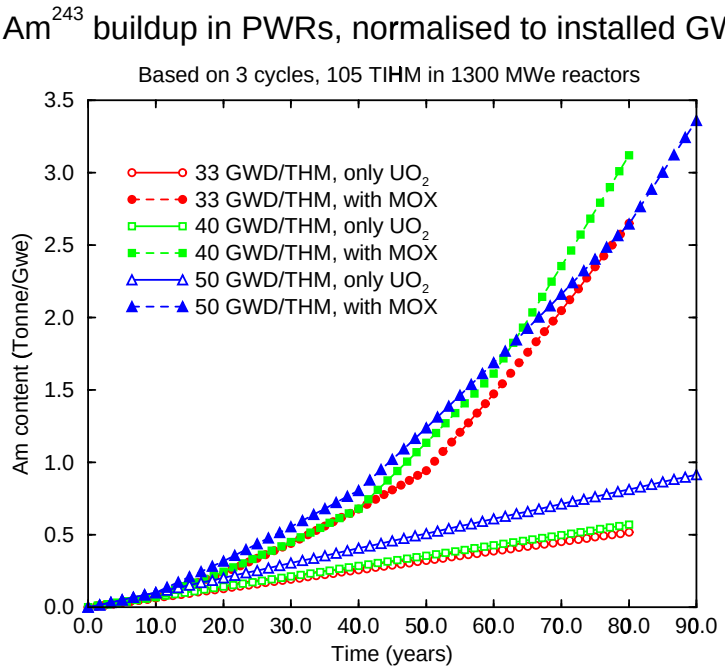


Figure 2: *Am*$^{243}$ buildup in PWRs for burnups of 33, 40 and 50 GWD/THM