TRANSURANICS TRANSMUTATION ON FERTILE AND INERT MATRIX LEAD-BISMUTH COOLED ADS

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Abstract

Different strategies for the back-end of the nuclear waste are explored, including different strategies of ADS application to nuclear waste transmutation. In this paper the results of the detailed simulation studies of ADS systems, both with fertile (Th) and inert (Zr compounds) matrix fuels, but always with lead-bismuth coolant will be presented. In addition, several options are considered for the plutonium isotopes: direct burning in ADS together with the minor actinides, a separate partial burning in MOX LWR before its load to the ADS and intermediate solutions. Depending on the case, the studies are performed from two perspectives: the situation of the equilibrium of the fuel cycle and the approach to the equilibrium from the actual LWR discharge composition.

1. Introduction

CIEMAT is actively working on the evaluation of the possible roles of ADS systems on the nuclear waste management within a collaboration agreement with ENRESA, the Spanish enterprise responsible for the radioactive waste management. Different strategies for the back-end of the nuclear waste are explored, from direct disposal to different strategies of ADS application to nuclear waste transmutation.

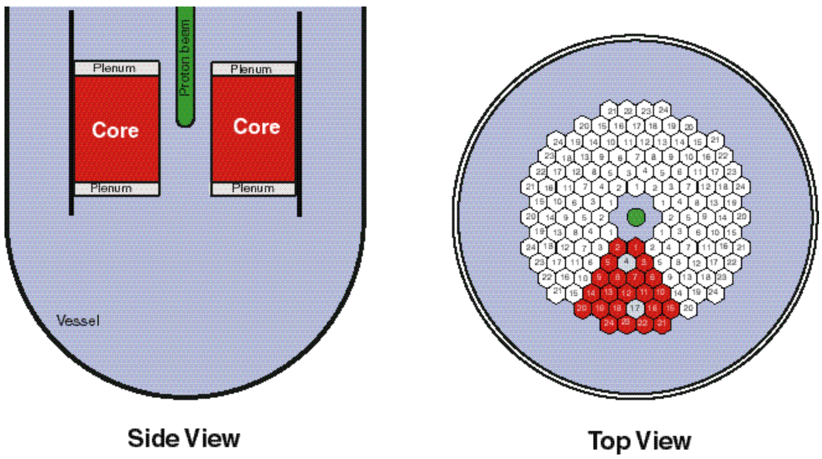
In this paper the results of the detailed simulation studies of ADS systems, both with fertile (Th) and Inert (Zr compounds) matrix fuels, but always with lead-bismuth coolant will be presented. In addition, several options are considered for the plutonium isotopes: direct burning in ADS together with the minor actinides, a separate partial burning in MOX LWR before its load to the ADS and intermediate solutions. Depending on the case, the studies are performed from two perspectives: the situation of the equilibrium of the fuel cycle and the approach to the equilibrium from the actual LWR discharge composition.

The studies are grouped in two wide groups. The first one is based on an ADS with a MOX fuel based on a ThO_2 matrix and the second one for the inert matrix cases is based on ZrN plus AcN. The ADS systems are similar for the two cases but not exactly the same, on the other hand the methodology of the detailed simulations is in both cases the same, and always based on the EVOLCODE system [1].

2. ADS systems main characteristics and simulation methodology

The ADS concept used in all the studies includes a fast core with an hexagonal arrangement of fuel elements cooled by lead (fertile matrix) or lead bismuth eutectic (inert matrix) in forced convection, and operates at constant thermal power close to 800 MW_{th}. The external neutrons are produced in a windowed spallation target, of the same material that the main coolant, by the action of a 1 GeV proton beam. The mass and composition of the fuel depends on the case.

Figure 1. **Side and top view of the ADS core concept used for the inert matrix simulations**



The ADS used for the fertile fuel case had already been presented in several papers and conferences [2,3]. In the inert matrix cases, the core geometry has been slightly modified, including a total of 132 fuel assemblies, to introduce 12 special rod positions (see Figure 1). These positions are reserved for control bars, shutdown bars, sample irradiation channels, special instrumentation and others, however in the present studies they have been considered as filled with coolant. Table 1 gives additional details on the inert matrix ADS concept.

Table 1. **Inert matrix ADS parameters**

Hexagonal fuel subassemblies		**Proton beam and spallation target**	
Flat to flat	210.96 mm	Kinetic energy	1 000 MeV
Total height	150 cm	Beam pipe material	HT9
Active length	120 cm	Beam window	Steel
Subassembly wall thickness	5 mm	Vacuum beam pipe thickness	3 mm
		Vacuum beam pipe external diameter	200 mm
Power + Primary circuit		**Fuel pins**	
Nominal power	800 MW_{th}	Number of pins per subassembly	Var. 169 – 331
Coolant/Convection type	Pb/Bi E./Forced	Pitch (mm)	Var. 15 – 10.7
Inlet temperature	300°C	External radius of fuel pins	4.1 mm
Outlet temperature	450°C	Cladding thickness	0.35 mm
Core		Void thickness	0.1 mm
Fuel	(Zr,TRU)N	External radius of fuel pellets	3.65 mm
TRU elements	Pu, Np, Am, Cm	Internal radius of fuel pellets	0.55 mm
Coolant and moderator	Pb/Bi		
Cladding material	Steel HT9		
Configuration	Hexagonal		
Number of fuel assemblies	132		
Number of special rod positions	12		

The simulation of the ADS systems, their k_{eff} values, power distributions and isotopic composition evolution during burn-up has been performed using the EVOLCODE system. The system is based on the combination of: LAHET [4] for the simulation of the neutron spallation in lead produced by the proton beam, and the transport of these neutrons down to 20 MeV; MCNP4B [5] for the complete neutron transport by Monte Carlo for energies below 20 MeV, and to calculate the neutron multiplication, the neutron flux energy spectra at different positions inside the core, the neutron flux intensity magnitude and distribution, the specific power distributions and the energy release by fission; and ORIGEN2.1 [6] with ad-hoc libraries for the burnup calculations. Further details on EVOLCODE can be found in [1]. For the purpose of the simulations of material evolution with burn-up, each fuel assembly is logically subdivided in 10 longitudinal zones.

3. Transmutation based on fertile or inert matrix ADS

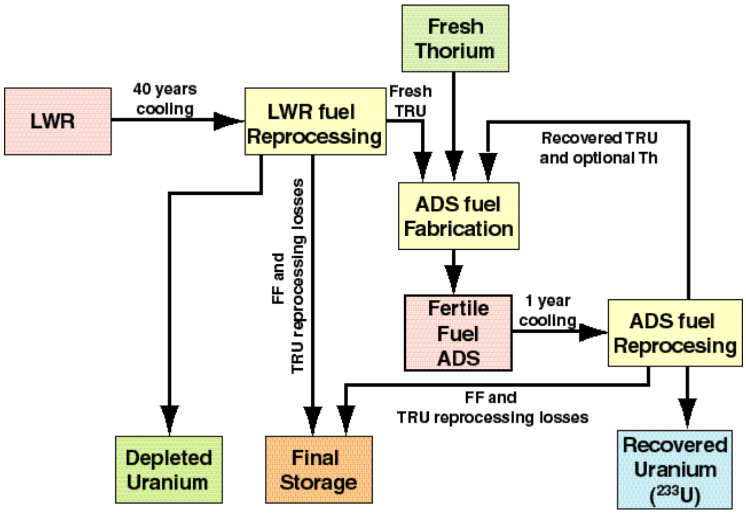
Two approaches are considered in the CIEMAT transmutation studies. The first one uses a Th matrix (ThO_2) for the fuel. The matrix provides chemical, mechanical and thermal characteristics very similar to the well known MOX fuels, and in addition, the breeding required to achieve very long burn-ups of the fuel (1 500 days). On the other hand, at the end of the transmutation process a substantial amount of ^{233}U has been bred from the Th matrix. This fuel cycle concept will make sense if the ^{233}U is used in the LWR substituting the ^{235}U or if the U-Pu cycle was to be replaced by the Th-U cycle. This second option will provide a much smaller production of transuranics and finally the radiotoxicity to be managed could also be reduced. In this last approach the use of TRU from the LWR in the ADS will be a transitory operation where most probably the equilibrium of the cycle will not be reached before the TRU are exhausted (depending on the different countries strategies). The study will concentrate on the first cycles of the TRU burning on ADS.

The most recent studies are devoted to evaluate the inert matrix option both in the mixed oxides or mixed nitrides versions. This option has the advantage of not introducing new isotopes in the fuel cycles, although the enrichment on some of the higher actinides becomes in certain phases unusually high. In the hypothesis of stability of the fraction of energy generation from the fission process, either the present LWR or new reactor types should provide most of energy and the ADS will only contribute with a small fraction of the total produced energy. In this circumstances the transmutation ADS has to handle a continuous amount of TRU regularly being produced at the same rate that they are eliminated. As in most strategies of transmutation on ADS, fuel recycling inside the ADS is required to obtain high elimination levels. It can be easily demonstrated that these two conditions are sufficient to progressively approach in the ADS to an equilibrium fuel composition. The behaviour of the system after reaching equilibrium decides the final TRU elimination efficiency of the system. For these reasons the studies of inert matrix concentrate on the fuel cycle after the equilibrium has been reached.

4. Fertile matrix (Th based) fuel option for TRU elimination

The concept explored in this study has been to close the LWR fuel cycle operation by introducing all the transuranic isotopes, TRU, contained in the LWR nuclear wastes, after 40 years cooling time, homogeneously in a fuel based on a thorium matrix. This fuel in the MOX chemical form is used in a fast ADS using lead as coolant. The ADS is then operated for a total of 1 500 days at a mean power of 800 MW_{th} reaching a burn-up of 146 GWd/THM (the burn-up for TRU reaches 238 GWd/T). The right choice of TRU/Th allows to obtain these very long burn-ups without interruption of the ADS operation, by the precise compensation of fissile isotopes consumption and breeding (mainly from the Th matrix). The ADS fuel is reprocessed after discharge, assuming an uniform 99.9% efficiency for all actinides. The fission and activation products and the reprocessing losses are stored in an appropriated repository. The uranium recovered, mainly ^{233}U, is available to be used in the operation of other reactors or ADS systems devoted to energy production. The recovered TRUs are mixed with fresh thorium and new TRUs from the LWR nuclear wastes to produce the new cycle fuel. Figure 2 and [2,3] provide more details of the global fuel cycle.

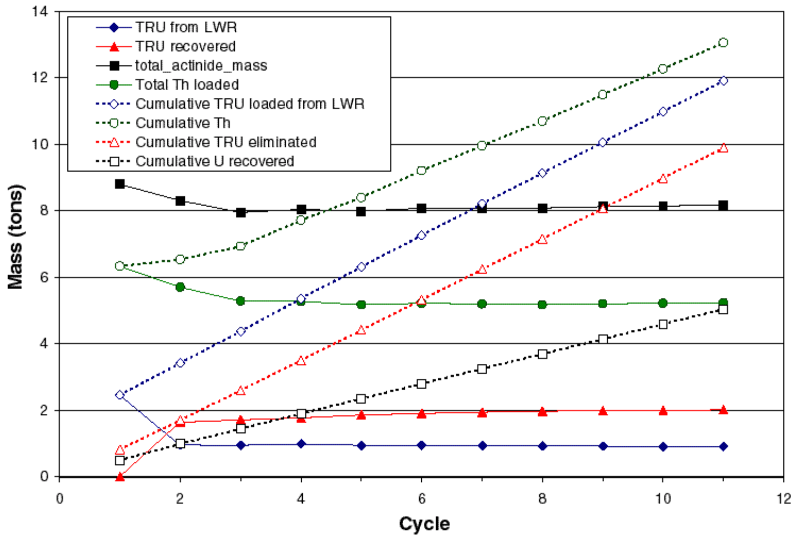
Figure 2. **Fuel cycle assumed in the fertile fuel ADS TRU transmutation studies**



The first eleven cycles of operation of such an ADS system had been carefully studied in detail. This number of cycles might be sufficient to exhaust most of the TRU contained in the nuclear wastes produced by one reactor generation (from beginning of nuclear reactors till the end of life of the presently installed LWR) of a country with moderate nuclear energy production (10 GWe) in a small number of ADS systems. The fuel composition of each reload is carefully tuned in order to maintain a sufficiently stable neutron multiplication (between 0.96 and 0.98) and to optimise the transmutation efficiency. The respect of the thermomechanical limits of the fuel during the burn-up is also verified.

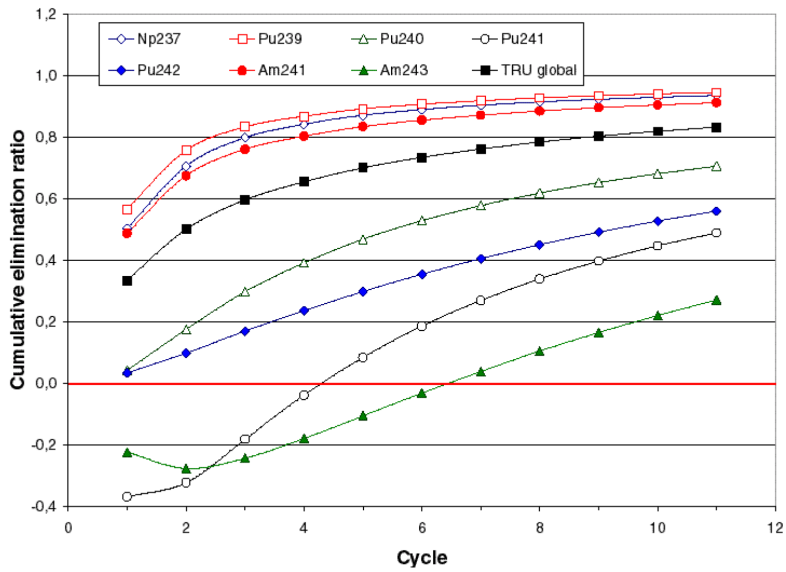
Figure 3 shows the masses of the different components of the fuel for each reload: the TRU recovered from the previous cycle, the new TRUs from the LWR wastes, the total Th and the total actinide mass. In addition, the figure also shows the accumulated TRU from LWR, accumulated Th entered in the system, the eliminated TRU and the recovered U after each cycle. It can be observed that these last four quantities increase linearly with the cycle number after the 4th cycle. The amount of TRU remaining in the ADS after each cycle decreases progressively approaching a constant value, as a consequence that as more and more cycles are performed and equilibrium is approached, the TRU transmutation efficiency increases reaching at the latest cycles very high values. The total TRU mass loaded in the eleven cycles was 11.9 tons while at the discharge of the 11th cycle the total remaining TRU is close to 2.0 tons. This means a global cumulative elimination ratio close to 83.1%. When reprocessing losses are taking into account the global cumulative TRU elimination efficiency is 82.8% in 11 cycles.

Figure 3. **Mass composition of the different ADS reloads and evolution of cycle parameters**



In addition to the mass reduction, the isotopic composition changes with the transmutation cycles. This is a consequence of the difference of the cumulative elimination efficiencies for the different isotopes. The evolution of this parameter for the most abundant TRU isotopes is presented in Figure 4. For all the main components of the LWR TRUs the cumulative elimination efficiency increases with the cycle number, reaching at the 11th cycle values as high as 94% for ^{239}Pu, 93% for ^{237}Np, 91% for ^{241}Am, 70% for ^{240}Pu, 49% for ^{241}Pu, 56% for ^{242}Pu and 27% for ^{243}Am. ^{238}Pu is not produced nor eliminated and the curium isotopes are continously produced in the system, but in any cases the final masses of these isotopes represent only 7% of the TRUs in the ADS discharge after the 11th cycle.

Figure 4. **Evolution of the cumulative elimination efficiencies for the most abundant TRU**



The theoretical limits of this system with very large number of cycles had been computed assuming that the 11th cycle is a good representative for the behaviour of the system at equilibrium.

Reprocessing losses both from the LWR and the ADS reprocessing had been taken into account assuming extraction efficiencies of 99.9% for all TRUs. The asymptotic limits of these curves had been computed giving as result that the 0.34% of the LWR TRU will end on the final storage, from reprocessing losses.

5. Inert matrix fuel options for TRU elimination

A parametric study of the characteristics of different inert matrix fuels with a ZrN matrix and different Pu-MA fractions [7] (from a cycle with UO_2 LWR and a single pass for all the Pu in LWR MOX fuel and in an ADS with 20 tons of nitride fuel), showed that the evolution of the ADS neutronic multiplication varies from a rapidly falling neutron multiplication to a continuous breeding to configurations close to critical as the MA fraction increases (see Figure 5). Of particular relevance is the existence of Pu-MA mixtures that allow achieving very long burn-ups with minimum variation of the neutron multiplication during the ADS operation. It is also important to note that the fraction Pu/TRU in these stable mixtures is approximately 40%, half the fractions produced in OU_2 LWR or in similar cycles.

The detailed studies on inert matrix fast ADS applications to nuclear wastes elimination had been performed on the scope of the cycle described in Figure 6. The UO_2 fuel is consumed in LWR. The resulting spent fuel is reprocessed separating four streams: the recovered depleted U, the Pu, the minor actinides (MA), and the fission fragments, activation products and reprocessing losses. The Pu is used to produce MOX and then this fuel is used once in LWR. The Pu and MA in the spent MOX and the MA from the LWR are send to the ADS described in Figure 1 and Table 1. The spent fuel of the ADS is then continuously recycled after reprocessing and addition of more Pu and MA from the spent UO_2 and MOX from the LWR.

Figure 5. **Evolution of the k_{eff} for different inert matrix Pu-MA fuel mixtures in a fast Pb-Bi cooled ADS**

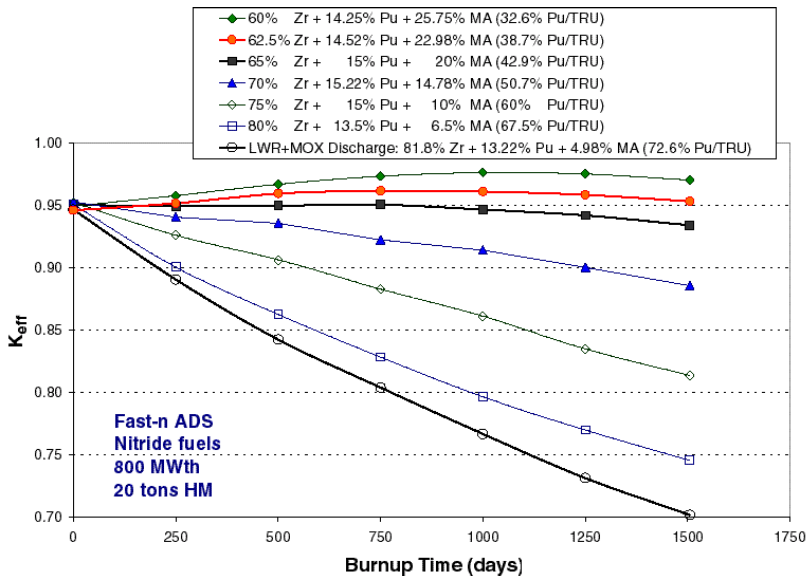
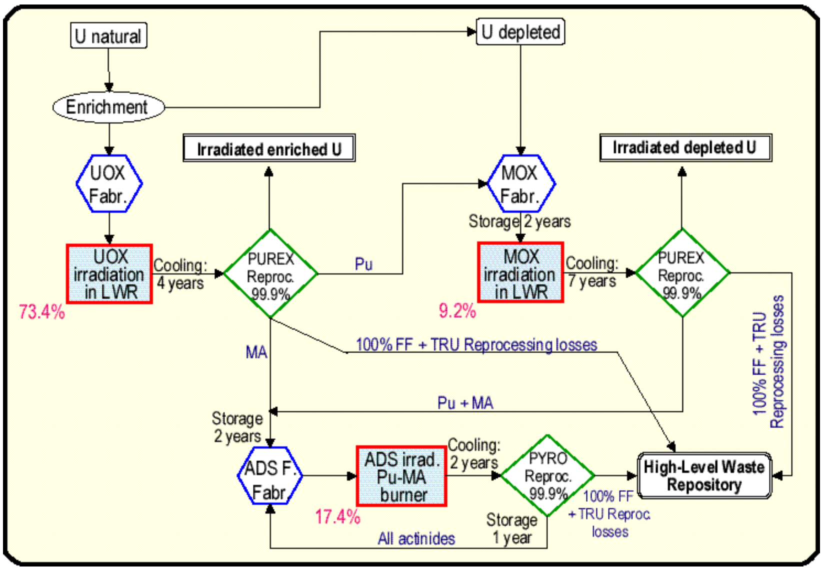
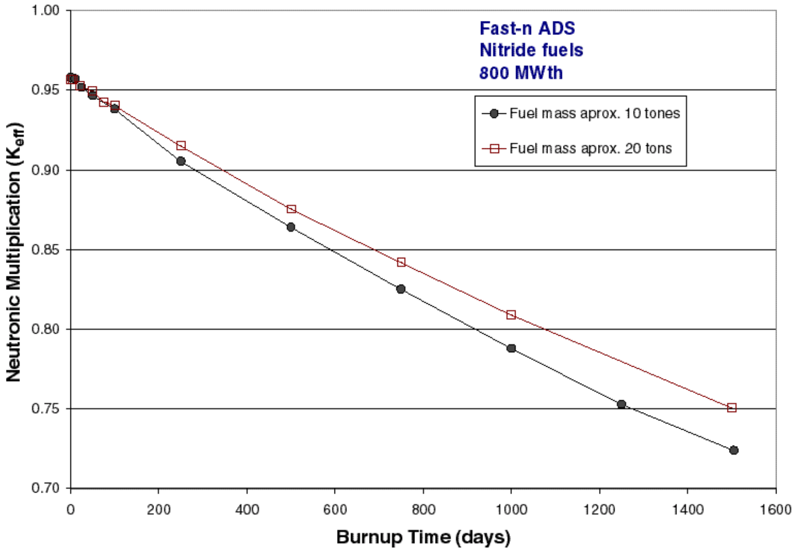


Figure 6. **Fuel cycle assumed in the inert matrix ADS TRU transmutation studies**



Because the transformation of the actinide isotopic composition vector along the ADS cycle (ADS burn-up, cooling, reprocessing and storage) is contractive, the fuel composition at the beginning of irradiation will progressively approach to an equilibrium value from cycle to cycle as far as the feed from the LWR reprocessing is kept constant. For the long-term consideration and in the hypothesis of maintenance of the present level of energy production from fission, the relevant information is the performance of the ADS cycle after this equilibrium has been reached. The isotopic composition of this equilibrium ADS fuel depend on the isotope vector coming from the ADS, the ADS characteristics and the burn-up per ADS cycle. This last dependence is however small and variations in the burn-up from 600 to 1 500 days introduce corrections smaller than 15% in the main isotopes. On the other hand the ratio between the LWR (UO_2 and MOX) TRUs and the total fuel mass in the ADS fuel depends strongly on this burn-up. The ADS equilibrium fuel composition was computed for the cycle of Figure 6. This fuel includes 2.5% U, 3.8% Np, 72.8% Pu, 13.1% Am and 7.8% Cm. The operativity of the ADS loaded with this fuel in nitride form distributed in a ZrN matrix was studied. Figure 7 shows the evolution of the neutron multiplication with the burn-up, for two different fuel masses of the ADS, 10 and 20 tons. The figure shows that it will be very difficult and expensive (from the accelerator point of view) to maintain the operation more than 150 days.

Figure 7: **Evolution of k_{eff} of an inert matrix fast ADS loaded with the equilibrium fuel of the cycle of Figure 6**



This peculiarity of the ADS loaded with this fuel will be a serious difficulty. On one hand, it will require frequent interruptions of the ADS that will reduce its energy production cost competitivity, and on the other hand, it will mean many reprocessing passes for the TRU before it is significantly reduced. Many possibilities can be envisaged to mitigate this difficulty in the ADS application to the transmutation of TRUs in equilibrium with simple LWR energy producing cycles. One type of possibilities already proposed by other authors are: the continuous or quasi-continuous fresh fuel supply by means of liquid fuels (e.g. molten salts), particle fuels (e.g. pebble-bed fuels), sliding fuel assemblies or designs of cores that allows to move fresh and spent fuel assemblies between the ADS core and a region neutronically decoupled inside the main vessel. A different possibility is the use of burnable absorbers or control rods in order to maintain a stable sub-criticality level. A third option, implicitly included in the double strata concept, consist in changing the isotopic composition of the equilibrium fuel. What is need is to severely reduce the Pu content on the equilibrium fuel. This can be achieved by reducing the Pu from the LWR reprocessing. This plutonium can not be simply stored, the natural option should be to use its potential as energy producing fuel by continuously reprocessing it on (critical or sub-critical/LWR or fast) reactors devoted to energy production. In this paper two additional options are discussed: the use of the equilibrium fuel in several batches and the use of a partially fertile matrix for the ADS.

5.1 Inert matrix fuel ADS for TRU elimination: batches with equilibrium fuel

One possible method to extend the burn-up of the fuel in the case of equilibrium fuel in an inert matrix ADS configuration is to irradiate the fuel in batches. Figure 8, shows a sketch of the possible batch refueling scheme studied in this paper. The approach is OUT-IN, with the fresh fuel coming to the ADS periphery there the fuel is irradiated for a period of time (166 days). When the neutron multiplication has fall below the accelerator possibilities, the ADS stops and the fuel elements move inward, extracting the inner most batch and introducing again fresh fuel in the periphery. Figure 9 shows the variation of neutron multiplication constant k_{eff} during one refueling bath. An equilibrium load has been computed that allows to charge exactly the same amount of fuel per 166 days batch in the 4 batches scheme, allowing to achieve a fuel burn-up at discharge of 140 GWd/THM, with a fluctuation on the k_{eff} from 0.956 to 0.936 during each batch. The fresh fuel composition introduced at the periphery has 75.3% Zr, and 24.7% of TRUs, with their equilibrium composition. This OUT-IN scheme allows also reducing the picking ratio of the power distribution inside the ADS.

Figure 8. **OUT-IN refueling scheme studied for the inert matrix ADS with equilibrium fuel**

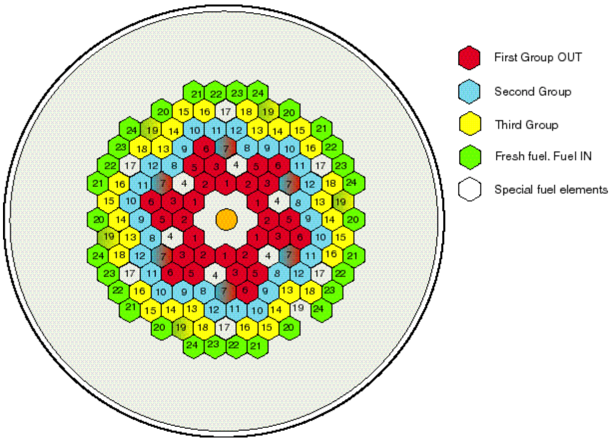
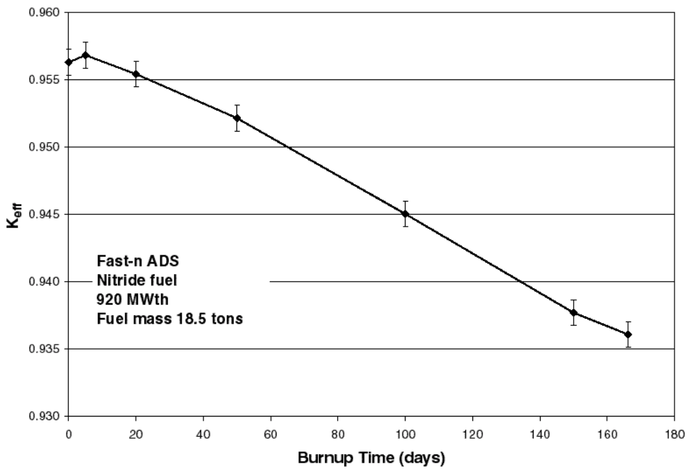


Figure 9. **k_{eff} evolution of an inert matrix ADS with equilibrium fuel during one of the four refueling batches**



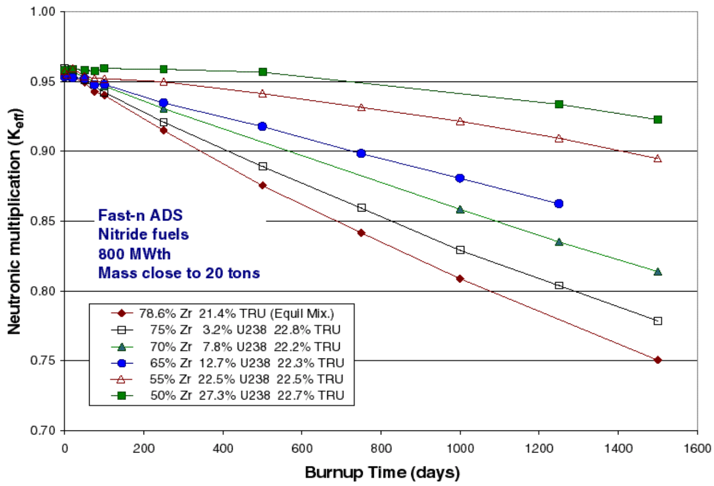
5.2 Partially fertile matrix fuel ADS for TRU elimination with equilibrium fuel

Another option to extend the burn-up of the fuel per ADS cycle is the use of partially fertile matrix adding either ^{238}U or ^{232}Th. The peculiarities of fertile fuel with Th had been described in the previous section. The natural choice for the breeding material should be ^{238}U. This isotope will introduce no new isotope in the fuel cycle and the main effect would be the reduction of transmutation efficiency per cycle. Figure 10 shows that a fuel with 65% Zr, 12.7% U and 22.3% TRU allows to extend the operativity of the ADS loaded with 20 tons of nitride fuel for more than 500 days. Configurations with 55% Zr, 22.5% U and 22.5% TRU and 21.7 tons of nitride fuel allow to operate for more than 1 200 days. Mixtures of Th with 70% Zr, 7% U and 23% TRU and 18 tons of fuel also allow to obtain operation for more than 500 days. These configurations improve the achievable burn-up per cycle of the ADS but reduce the transmutation efficiency of TRU. Figure 11 shows the change in TRU elimination from pure inert matrix to the 65% Zr, 13% U and 22% TRU fuel after 500 days of irradiation. The eliminated TRU mass is 72% of what could be transmuted in a pure inert matrix ADS if it could be operated for 500 days. The main effect of this reduction is to increase in the complementary proportion the number of reprocessing passes and the corresponding reprocessing losses, as well as increasing the time required for TRU elimination. Both inconveniences are easily acceptable and well compensated for the extension of the single pass burn-up.

5.3 Reprocessing losses estimation

To estimate the TRU fraction finally going to the nuclear waste storage, from the reprocessing losses, in the inert matrix scenario, the 4 batches refueling concept with 660 total irradiation time and an average burn-up of 140 GWd/THM, will be used, as the simpler solution for a realistic operation of an inert matrix TRU transmuter ADS. For these parameters and assuming that the reprocessing efficiencies are 99.9% for all the TRUs in the reprocessing of the LWR, the MOX and the ADS spent fuels, simple arithmetic allows to estimate the fraction of TRUs going to the repository between 0.7 and 0.8% of the originally produced. The value obtained from the detailed simulation is 0.707%.

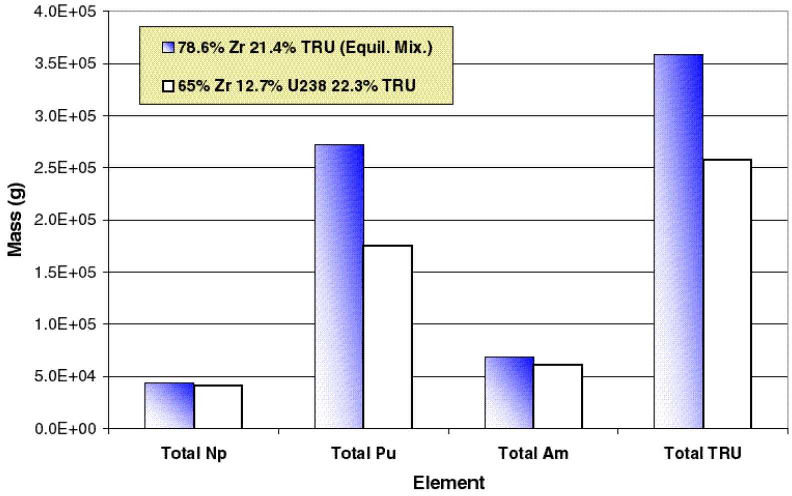
Figure 10. **k_{eff} evolution of an partially fertile (Zr-^{238}U) matrix ADS with equilibrium fuel**



6. Conclusions

The previous exercises have shown that both the inert matrix and fertile matrix allow to reduce the amount of TRUs to be stored in the final nuclear waste repository by a factor larger than 100 if the cycle is maintained sufficiently long. The inert matrix choice is the solution of minimum perturbation of the present fuel cycle but it has the difficulty of short burn-up per cycle. Several solutions are possible for this problem, again the minimum deviation from the present cycle would be the use of refueling batches or partially fertile (Zr-^{238}U) matrixes. A more advance solution is the introduction of Pu recycling in the energy production strata, although this probably will require the use of new types of reactors. Finally the use of Th based matrix ADS will be more justified as a transition from the U-Pu fuel cycle to the Th-U fuel cycle for energy production, although intermediate solutions are also possible.

Figure 11. **Transmutation efficiency per 500 days batch in inert and partially fertile matrix ADS**



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