

NATIONAL TRANSMUTATION STRATEGIES OF GERMAN SPENT FUEL LEGACY TO REDUCE THE IMPACT ON DEEP REPOSITORY

A. Schwenk-Ferrero¹⁾, J. Knebel²⁾, W. Maschek¹⁾

Forschungszentrum Karlsruhe GmbH, Technik und Umwelt, Postfach 3640, 76021 Karlsruhe, Germany

¹⁾ *Institute for Nuclear and Energy Technologies, Aleksandra.Schwenk-Ferrero@iket.fzk.de*

²⁾ *Programme Nuclear Safety Research*

The major objective of P&T is to alleviate the burden on the geological disposal. Transmutation performance should be analyzed however in a broader context by considering top-level goals that must be reached by a complete fuel cycle associated with a particular transmutation strategy including improved public safety, benefits to repository program, reduction of proliferation risk and improved prospects for nuclear power. NEA/OECD launched 2002-2005 expert studies on the impact of advanced fuel cycles on waste management policies applying a set of basic indicators like waste volumes, resource consumption, radiation levels, thermal decay heat or actinide content to judge whether the top-level goals can be achieved by a candidate transmutation system. Here studies for German P&T strategies in the framework of national R&D program evaluating the performance of feasible "industrial scenarios" are discussed. Two representative fuel cycle schemes: current technology and fully closed, acceleration-driven fuel cycle were investigated. For each class mass flow, waste generation (the waste being classified according to categories defined by IAEA as short-lived waste for surface or sub-surface disposal, long-lived, low heat producing and high-level waste for geological disposal), repository performance are assessed and some economic issues of ADS deployment are addressed.

I. INTRODUCTION

In exploring different partitioning and transmutation (P&T) options an integrated nuclear waste management strategy is a critical issue, inevitable for the long term sustainability and growth of nuclear power. The primary objective of P&T strategy is thereby to reduce the amount of transuranics and by this the long-term radiotoxicity and heat load of the high-level waste (HLW) going to deep geological repository. Candidate P&T approaches which implement innovative fuel cycle schemes were extensively investigated with respect to their impact on waste repository performance by dedicated US and NEA/OECD expert teams^{1, 2)}. Overall findings of these

investigations show that it is possible to establish a strategic progression towards a maximal reduction of waste volume to be disposed off and maximal reduction of specific uranium consumption. Moreover they bring into evidence the achievable reduction of waste activity and heat load while burning plutonium and minor actinides in dedicated critical and/or accelerator driven systems. Apart from repository benefits, the transmutation performance should be evaluated against other top-level goals such as improved public safety, reduction of proliferation risk and improved prospects for nuclear power. A set of representative indicators or specific programmatic criteria has already been established to facilitate the evaluation of system performance for candidate approaches.

In both US and OECD studies multi-tier transmutation systems were considered, nine representative fuel cycle schemes were investigated which belong to three families: schemes based on current industrial technology and possible extensions, schemes with partially closed fuel cycles and schemes involving fully-closed cycles deploying fast reactors and ADS based transmuters. The analyses were performed for nuclear systems in equilibrium because the fuels and processes (reprocessing and fabrication) considered in the studies – and hence the resulting waste streams - are not sensitive to transient phenomena.

The US study focuses primarily on national circumstances whereas the OECD study addresses mainly expanding nuclear economies worldwide. In particular of countries which have ongoing commitments to use MOX fuel, might move toward infinite recycling of MOX, implementing MOX-UE schemes and might deploy in the future Gen-IV or subcritical systems pertaining to the generation of power, stabilizing plutonium resources and transmuting accumulated MA stocks. Both studies provide a vital policy guidance showing whether the mature state of the proposed nuclear economy is a desirable one. They cannot take, however, into account the real-world initial conditions and time dependent variations of deployment strategies, nor do they account

for the time taken to move from the current reactor fleet configuration to the equilibrium state.

For Germany, which is phasing-out the nuclear power and in the near future has to manage its spent fuel legacy, P&T strategy based on the accelerator-driven systems might offer an attractive option to reduce plutonium (Pu) and minor actinides (MA) inventories. Giving the strongly time-dependent nature of German nuclear economy it is desirable to look beyond a static or quasi-equilibrium paradigm when considering the course that might be taken by nuclear power. For this purpose use was made of nuclear fuel cycle analysis code NFCSim which was developed in LANL³⁾.

II. PARTITIONING AND ADS-BASED TRANSMUTATION

In the framework of the national R&D program feasible “industrial P&T scenarios” were explored with the NFCSim code⁴⁾ and their performances were evaluated against a relevant subset of goals, indicators and criteria similar to those^{1, 2)} applied in the international studies. It was postulated that Germany addresses its Spent Nuclear Fuel (SNF) inventory by pursuing an ADS based P&T strategy. A prerequisite for this study was an assessment of the final spent nuclear fuel inventory when the German nuclear fleet is retired. To assess the stockpiles of Pu and MA in the SNF, the case of a progressive phase-out of the nuclear power fleet of Germany, presently foreseen in 2022, was considered. The German fleet, 13 Pressurized Water Reactors (PWR) and 6 Boiling Water Reactors (BWR), some of them (~60%) fuelled with uranium and plutonium mixed oxide fuel (MOX), provided in 2002 the approximate electric power of 20 GWe. The NFCSim prediction of the evolution of the SNF inventory for this fleet is given in Table I. The calculations were performed accounting for an early cessation (~2005) of the SNF reprocessing.

Table I. Inventories (tons) of German SNF and HLW as of January 1, 2022

Quantity	PWR UOX	PWR MOX	BWR UOX	BWR MOX	Tot. SF	HLW
Total	5350	773	3470	246	9840	2.15E+02
U	5060	702	3310	227	9290	6.64E-01
Pu	51.7	34.3	32.9	7.95	127	2.01E-01
Np	3.6	0.234	2.16	0.0497	6.04	2.94E+00
Am	4.6	4.96	3.48	1.17	14.2	3.63E+00
Cm	0.23	0.226	0.148	0.0644	0.669	7.36E-02

In 2022, the SNF will contain 127 tons of plutonium. In the vitrified HLW, the bulk of the mass (~96.7%) is contributed from the fission products. The trace actinides follow from the assumed 99.8% recovery efficiency of all transuranics (TRU).

Figure 1 shows the evolution from 1970 to 2022 of the existing SNF inventory and the cumulative spent fuel reprocessed in Germany. By 2022, 16840 tHM will have been discharged from German reactors.

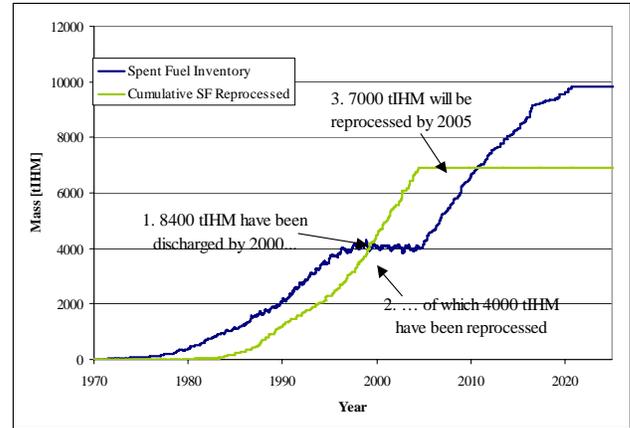


Fig. 1. Spent fuel inventory and integrated reprocessing throughput for the German fleet.

Reprocessing and utilization of recovered plutonium in MOX fuel reduces the heavy metal to be finally disposed off to 9840 tons, i.e. about 42%.

Further on, it was investigated to which degree ADSs could contribute to mitigating the burden of SNF disposal in Germany. The ADSs will be deployed beginning in 2030. The ADS park will be so sized that all German SNF will be reprocessed during the 40 year lifetime of the ADSs. Subsequently, a smaller fleet of ‘second generation’ ADSs will be deployed as the first generation facilities retire. Hence, the simulation commences in 2030 and extends over approximately 100 years, about two facility lifetimes. The progress made in reducing actinide inventories in 2100 as well as upon retirement of this second generation is assessed. The ADS is a Na-cooled, metal-fuelled facility with an LBE target. Table II provides a summary of parameters used for this facility and its associated fuel cycle.

Table II. Top-Level ADS Design Parameters

Target k_{eff}	0.97 (BOC); 0.94 (EOC)
Core inventory	3000 kgIHM
Thermal power	840 MW _{th}
Discharge burnup	200 MWd/kg
Fuel management	5 batches / core
Cycle time	168 days (142.9 efpd)

The ADS fleet size is determined by the amount of material available for transmutation: the fleet must be of sufficient size to take up, as nearly as possible, the entire SNF inventory during the lifetimes of the first generation of transmuters (40 years). Hence, eight 840 MW_{th} facilities were deployed in the first generation and three in the second since half of the TRU content in SNF was transmuted by the first generation. The deployment pace of ADS units is shown in Figure 2.

The annual oxide fuel reprocessing throughput is depicted in Figure 3. In this strategy, since plutonium constitutes ~85% of the TRU contained in SNF, the ADSs

used to transmute TRU must necessarily employ relatively short cycles.

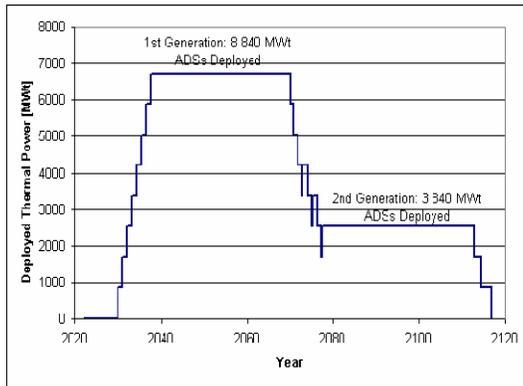


Fig. 2. ADS deployment schedule for transmutation of German SNF.

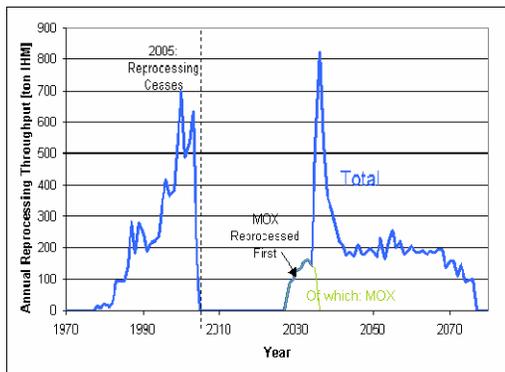


Fig. 3. Annual reprocessing throughput.

In fact, it was found that the steep burnup reactivity gradient resulting from using given TRU inventory limits the ADS cycle burnup to 40 MWd/kg (with a reactivity swing $\Delta k_{eff} = 0.03$) and the cycle time to slightly less than one half of a year.

III. IMPACT ON TRANSURANICS STOCKS AND REPOSITORY

Over two generations of ADS deployment Germany would transmute 82% of the 129 tons of plutonium and 45% of the 35.8 tons of MA it possesses in 2022. In fact, since 7.4 tons of MA has been already vitrified prior to 2005, it is better to state that Germany disposes off 57% of the 28.4 tons of MA present in SNF in 2022 and thus available for transmutation (see Figure 4).

To quantify the implications of this strategy on disposal options, the decay power of all nuclear material in the system was evaluated at several points in time. At any given time, this evaluation is carried out based upon all materials that have been out of pile for longer than five years. Younger SNF is discounted because the presence of

very short-lived nuclides would render the results difficult to interpret.

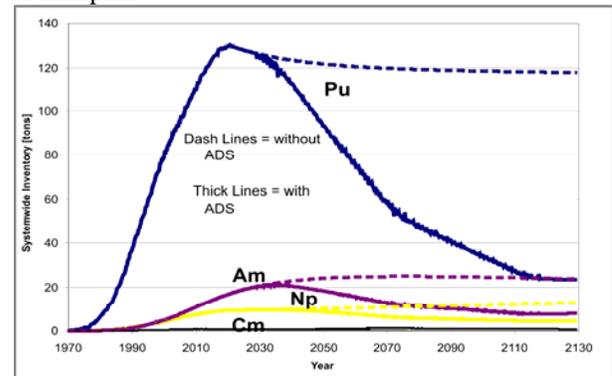


Fig. 4. The effect of ADS deployment on transuranics inventories.

The instantaneous decay power of the SNF and vitrified HLW is shown on Figure 5.

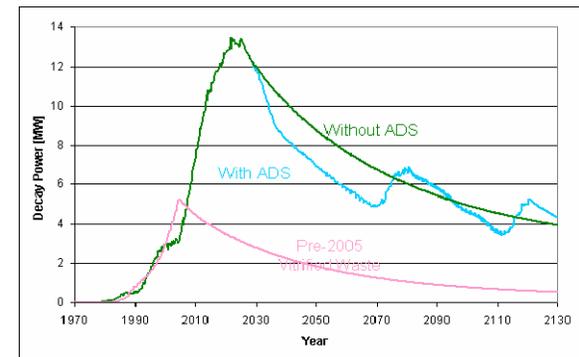


Fig. 5. Decay power of stored nuclear material at date shown.

It is apparent that the current strategy incorporating ADS transmutation diverges from the reference case in 2030. Increases in the decay power associated with the transmutation strategy after 2070 and 2110 are assigned to the shutdown of ADSs and discharge of their final cores.

It is of interest to observe that the short-term heat release rate of the oxide SNF is approximately the same as that of the HLW and spent metal fuel discharged from the ADSs. The bulk of the decline in the heat production rate of the oxide SNF during this time period is ascribed to the decay of Sr-90 and Cs-137. ADS transmutation would seem to offer little benefit in the very short term simply because Sr-90 and Cs-137 are continuously being created during the operation of the ADS fleet. This new influx of high heat release fission products offsets, in the very short term, the benefit gained from fissioning the transuranics.

The benefits of the transmutation strategy become apparent when one looks to heat production in the longer term. The decay power of stored nuclear material following 100 years of cooling is shown in Figure 6. In

this figure, the value given at, say, 2020 reflects the heat production rate of all material that is out of pile in 2020 evaluated at 2120. Assessed decay power constitutes a relevant metric for the long-term interim storage needs or for the early phases of repository operation, depending on the disposal strategy pursued. The benefits of transmutation are still partially offset by ongoing production of fresh nuclides with high heat release, but to a lesser extent than it was the case for the short-term decay heat. It is seen that two generations of transmutation reduce this medium-term heat load burden by roughly a factor of two.

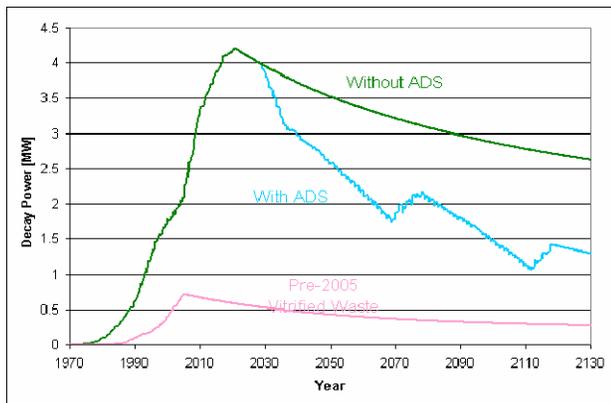


Fig. 6. Decay power of stored nuclear material 100 years after date shown.

The long term decay heat – the decay power integrated over a period extending from 100 to 2000 years in the future – is shown in Figure 7. Since transuranics, particularly Am-241 and Pu-238, dominate heat production on this time scale, destruction of most of these isotopes via transmutation is seen to offer a substantial benefit: the decay heat production is reduced by a factor of four following two generations of ADS operation.

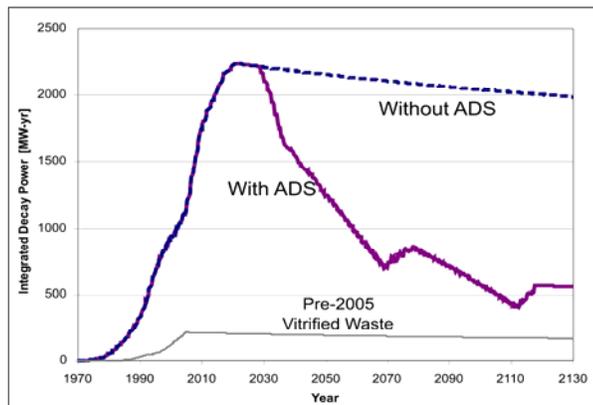


Fig. 7. Decay power of stored nuclear material, integrated over period from 100 to 2000 years after date shown.

Decay heat is a major input for the design of underground repositories. For disposal in granite, clay and

tuff formations the maximum allowable disposal density is determined by thermal limitations. HLW arising from ADS fuel cycle scheme generates considerably less heat than the spent fuel arising from the PWR or BWR one-through scheme or schemes with single recycle of separated plutonium in form of MOX fuel. This lower thermal output of HLW allows a significant reduction in the total length of disposal galleries needed. Separation of caesium and strontium would reduce further required repository size. For example, in the case of disposal in clay formation the gallery length needed in the HLW disposal can be reduced by a factor 3.5 through a fully close fuel cycle scheme as compared with the reference PWR once through scheme and by a factor 9 through a scheme including separation of caesium and strontium²⁾. In case of disposal in rock salt the heat generation of the disposed waste contributes to a fast salt creep and void volume reduction. Therefore, a lowering of the thermal output of the high level waste forms necessitates an optimization of the work packages and of the disposal configuration. The volume of HLW to dispose off is a driving factor to determine the load capacity of a given repository site. Differences in heat load and waste volume may have a major impact on the detailed concept of the repositories.

A transmuting fleet consisting of accelerator driven systems can thus significantly alter, and by most metrics reduce, the burden of spent fuel and waste disposal. Moreover, the evolved composition of plutonium present in SNF fulfils better non-proliferation criteria after two generations of transmutation⁴⁾. Table III shows the isotopic content of all plutonium present in SNF in 2022 and 2122. For simplicity, oxide SNF – PWR and BWR, UOX and MOX – is lumped. In 2022, the SNF is the mixture of UOX and MOX as given in Table I. In 2122, only ADS SNF is present. It is clear that the plutonium resident in ADS SNF is of little value for weaponization, even ignoring the substantial intrinsic radiation barrier to separations posed by the SNF itself.

Table III. Proliferation-Relevant Attributes of German Plutonium Vectors Averaged Over all SNF at Dates Given

	238 %	239 %	240 %	241 %	242 %	Decay Heat [W/kg]	Spont. Fission Neutrons [# /kg/s]	Bare Sphere Critical Mass [kg]
SNF in 2022	2.4	54.4	28.1	7.5	7.7	16.9	450,000	14.7
SNF in 2122	10.5	13.9	52.6	4.2	18.7	63.9	1,080,000	22.0

For radioactive waste management Germany adopts the waste categories defined in the IAEA classification shown in Table IV. IAEA classification system distinguishes radioactive waste types based on two key

characteristics: thermal hazard and requirement for disposal. Types of wastes identified are: high-level waste and low- and intermediate-level waste (LILW) with short (LILW-SL) or long life (LILW-LL) times. Germany intends to dispose off all types of radioactive waste in deep geological formations⁵⁷. This intension makes it unnecessary to differentiate between the waste containing radionuclides with comparatively short half-lives and waste containing radionuclides with comparatively long half-lives. As such, there are no measures or precautions required in Germany in order to separate LILW according to its life time. Thus, the basic German classification makes a subdivision only into heat-generating radioactive waste (HLW) and radioactive waste with negligible heat generation: low-level waste (LLW) together with intermediate-level waste (ILW). Waste with negligible heat generation consists of operational waste from nuclear power plants (e.g. filters, ion exchange resins, clothes or cleaning rags), decommissioning waste as well as radioactive waste from research, medicine and industry. Heat-generating waste consists especially of the vitrified fission product solution originating from reprocessing of spent fuel elements and SNF discharged from power plants envisaged for direct disposal. The management of SNF is restricted by law to direct disposal and presently obligates the nuclear power plant utilities to provide on-site interim storage facilities. Decentralized on-site facilities may either be the interim storage facilities with the operational lifetime of 40 years or the storage areas with the operational lifetime of 5 years, respectively. Commencing from 1998 till Feb. 2002, 18 applications were made by the utilities for dry on-site interim storage. Till 2005 all licenses were granted; three interim storage areas and six interim storage facilities were constructed and put in operation.

By the year 2080 German radioactive inventory from the current nuclear programme is expected to include approximately 290000 cubic meters of conditioned LILW and approximately 24000 m³ of conditioned HLW. Since Germany has confidence that geological disposal is the best end point for managing radioactive waste sites are nowadays being selected. Till now one site has been already used and two have been explored for the disposal. These are: Morsleben repository for LILW with mainly short lived radionuclides and alpha emitters concentration up to 4.0 E+11 Bq/m³, Konrad repository (former iron ore mine), investigated for the disposal of all short- lived and long-lived waste with negligible heat generation, and Gorleben repository. Waste with negligible heat generation comprises waste packages which do not increase the host rock temperature of Konrad by more than 3K on average. Originally, it was planned to put up in Konrad repository of up to 650000 m³ waste package volume with a total activity of about 1E+18 Bq for beta/gamma and about 1E+17 Bq for alpha emitters, respectively. Waste packages should be emplaced at a

depth from 800 to 1300 m in disposal cavities with a cross-section of 40 m² and a length of up to 1000 m by using a stacking technique. The licensing procedure for the Konrad repository was started on 31 August 1982.

Table IV. IAEA Recommendation for Qualitative Waste Classification

Category	HLW (deep geological disposal)	LILW-LL (geological disposal)	LILW-SL (surface or geological disposal)
Main characteristic	(i) Highly radioactive waste, containing mainly fission products, as well as some actinides, which is separated during reprocessing of irradiated fuel (ii) Spent fuel, if it is declared a waste.	(i) Waste which, because of its radionuclide content requires shielding but needs little or no provision for heat dissipation during its handling and transportation.	(i) Waste, which, because of its low radionuclide content, does not require shielding during normal handling and transportation.
Heat generation	(iii) Any other waste with radioactivity levels intense enough to generate heat more than 2 kW/m ³ by the radioactive decay process.	(ii) Heat generation < 2 kW/m ³ .	(ii) Heat generation < 2 kW/m ³ .
Half-life		(iii) Half-life > 30 a.	(iii) Half-life < 30 a.
Other characteristic			(iv) Activity content < 400Bq/g of long lived alpha emitters.

The license for the emplacement of up to 303000 m³ issued in May, 2002 was immediately claimed against. This caused the interruption of the preparations for radioactive waste put up until the judicial decision becomes effective. Gorleben repository project has been started late seventies. Gorleben salt dome was extensively investigated for its suitability to host an underground repository at depths between 840 m and 1200 m for all type of radioactive waste, in particular HLW. The accumulated inventory of beta/gamma and alpha emitters should be in the order of magnitude of 1E+21 Bq and 1E+19 Bq, respectively. As soon as the programme of the site specific investigations has been accomplished two shafts were completed. In 1998, however, a decision was made to review the German waste management concept. At present, various sites with different host rocks are under investigation and finally these sites will be chosen which perform best in terms of security, as well as protecting the public and the environment.

The estimation for the amount of LLW, ILW and conditioned HLW generated in different fuel cycles (FC) can be found in the NEA/OECD study²⁾. Figures 8-10 plot relevant data extracted from OECD assessments.

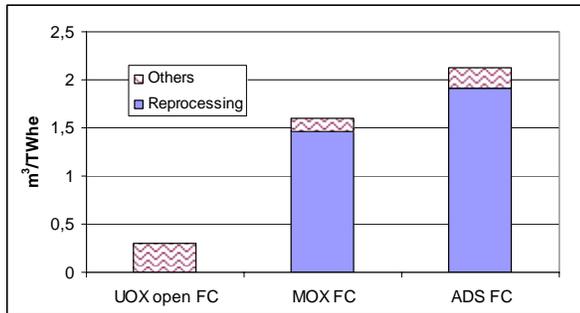


Fig. 8. Total amount of long-lived LILW generated in FC

The amount of LILW-LL generated in the fuel cycles depends on the spent fuel reprocessing technology. Regarding the reprocessing of fuels from dedicated ADSs actinide burners charged with nitrite fuel, the activity of discharged burn-up fuel is so high that the radiation stability of the organic extractant in aqueous processes can no longer be guaranteed and the fuels can only be reprocessed by means of pyrochemical methods. The pyrochemical methods produce in turn new types of waste streams and waste forms which have not yet been characterized fully. The data suggests, as natural, that each additional separation step produces slightly more waste. Thereby, the data concerning ADS fuel cycle scheme should be interpreted with a great care.

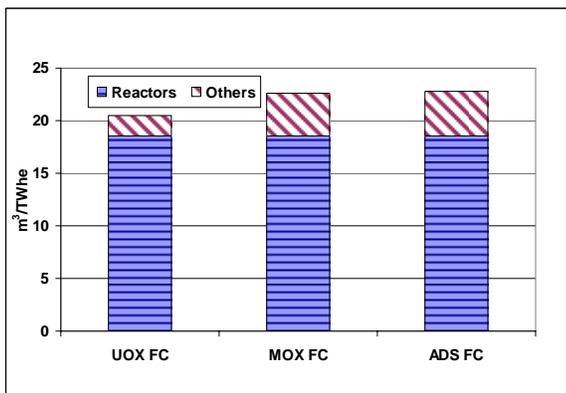


Fig. 9. Total amount of short-lived LILW generated in FC

The waste volume depends on conditioning technology and the values presented in Ref. 2. must be seen as based on the preliminary estimates.

The volume of the HLW is largely defined by the waste loading factor when the waste is conditioned. In many cases the limiting factor is the amount of fission products. Thus, the amount of actinides in the waste has only a minor effect on the HLW volume. The volume of

HLW of once-through cycle corresponds to the volume of fuel element. Thus, the density of the waste is much lower as compared to the conditioned HLW coming from reprocessing. Total amount of conditioned HLW for three fuel cycle schemes is depicted in Figure 10.

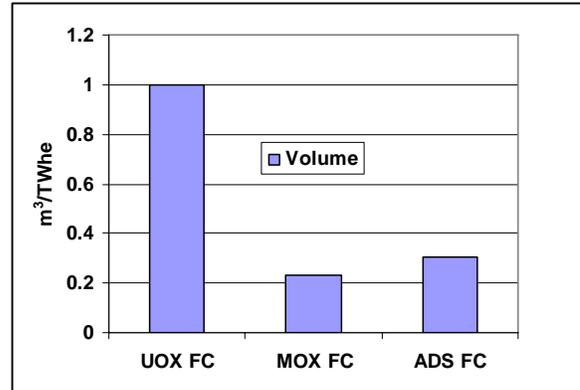


Fig. 10. Total amount of conditioned HLW

According to Ref. 2, the volume of conditioned waste between different categories decreases roughly by factor of 10 and amounts (per TWhe) to 20-25 m³ for LILW-SL, 1.5- 3 m³ for LILW-LL and 0.12-0.4 m³ for HLW.

IV. RELATIVE COST OF TRANSMUTING STRATEGY

Each program implementing the P&T strategy would possess three components: R&D, pilot facility deployment and operations. A detailed cost study of such program has been conducted for the case of deployment of 64 840 MW_{th} piles for treatment of US SNF⁶⁾. Although neither this program nor the facilities considered are perfectly analogous to the scenario investigated herein, the cost and scheduling data given for ATW system are sufficient to allow the ADS system life cycle costs and revenues associated with German P&T scenario to be approximated well. For this purpose, a simplified version of the ATW cost spreadsheet presented in Ref. 6 was prepared⁷⁾. The ADS deployment scheduling was then imposed.

It was found that many factors combine to make an estimate of the absolute system costs drawn from the data mentioned above questionable, for instance:

- In Ref. 6 significant resources development and deployment of modular, integrated LWR fuel reprocessing capability are assigned. It is not clear that this would be desirable in Germany, where the industry is already mature.
- A 60 year facility lifetime is used, whereas German study assumes 40 years.
- The ADSs themselves are similar but not identical:

just one difference, a larger allowed k_{eff} swing, might lead to an as yet unclear effect on construction costs, a decrease in the power delivered to the grid and an increase in the load factor.

- The costs used reflect US conditions. A subtle example of this is found by considering, for instance, costs associated with the metal fuel fabrication facility. These were derived based upon the TRU inputs relevant to the proposed US ADS park; it is not clear that they remain valid for the (generally hotter) feed streams associated with the P&T scenarios considered here.

The issues raised above largely vanish if one limits oneself to studying the relative costs associated with ADS transmuting strategy. Annual cost and revenue were computed, in year 1999 US dollars, for two generations of facilities (life cycle costs are tabulated for all ADSs constructed prior to 2100). Revenue was calculated assuming that the R&D program begins in 2005 and large-scale deployment in 2030 and that the electricity can be sold at 4.3 cents / kWh. Figure 11 shows the estimated cash flows for Germany ⁷⁾.

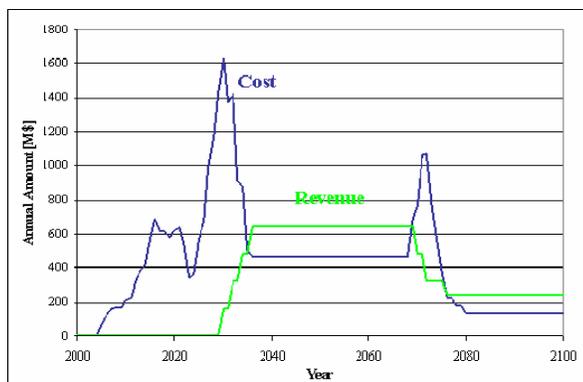


Fig. 11. Undiscounted Annual Cash Flow for Germany

V. CONCLUSIONS

The implementation of P&T strategy in Germany would result in a fivefold reduction in plutonium inventories over two generations of ADS operation. Furthermore, the MA mass present in SNF in 2022 and available for transmutation could be reduced by 57%. As compared to the no-action alternative, these accomplishments reduce the medium- and long-term heat production of the waste inventory by 50% and 72% respectively. NFCSim results also show large reductions in long-term radiotoxicity: the inhalation toxicity after 10,000 years is reduced by 79% and the ingestion toxicity by 76%.

Against these gains must be set the cost associated with deployment of 11 ADS plus oxide fuel reprocessing and dedicated metal fuel fabrication and reprocessing infrastructure.

ACKNOWLEDGMENTS

Part of this study was performed in collaboration with M. Salvatores (CEA), E. Schneider (formerly affiliated to LANL) and H.W. Wiese (FZK). NFCSim code developed at LANL was used as software tool to simulate the fuel cycles.

REFERENCES

1. G. J. VAN TUYLE et. al., "Candidate Approaches for an Integrated Nuclear waste Management Strategy – Scoping Evaluations", Los Alamos National Laboratory Report LA-UR-01-5572 (2001).
2. OECD Nuclear Energy Agency, "Advanced Nuclear Fuel Cycles and Radioactive Waste Management", NEA No. 5990 (2006).
3. E. SCHNEIDER et. al., "NFCSim: A Dynamic Fuel Burn-up and Fuel Cycle Simulation Tool", *Nuclear Technology*, **151**, 35 (2005).
4. E. SCHNEIDER, M. SALVATORES, A. SCHWENK-FERRERO et. al., "NFCSim Scenario Studies of German and European Fleets", LANL Report, LA-UR-04-4911 (2004).
5. R. WERNIKE, "The Control of Safety of Radioactive Waste Management and Decommissioning in Germany", OECD, NEA (2006) <http://www.nea.fr/html/rwm/rf/germany.pdf>.
6. R. I. SMITH et. al., "Estimated Cost of an ATW System", Pacific Northwest National Laboratory Report PNNL-13018 (1999).
7. E. SCHNEIDER, Los Alamos National Laboratory, personal communication (2004).