Hindawi Publishing Corporation
Science and Technology of Nuclear Installations
Volume 2008, Article ID 756181, 6 pages
doi:10.1155/2008/756181

Research Article

Wastes Management Through Transmutation in an ADS Reactor

Barbara Calgaro,[1] Barbara Vezzoni,[1] Nicola Cerullo,[1] Giuseppe Forasassi,[1] and Bernard Verboomen[2]

[1] *DIMNP, Faculty of Engineering, University of Pisa, Via Diotisalvi n.2, 56122 Pisa, Italy*
[2] *Advanced Nuclear Systems Institute, SCK·CEN, Boeretang 200, 2400 Mol, Belgium*

Correspondence should be addressed to Barbara Calgaro, barbaracalgaro@gmail.com

Received 26 June 2007; Revised 29 December 2007; Accepted 7 April 2008

Recommended by Giuseppe Vella

The main challenge in nuclear fuel cycle closure is the reduction of the potential radiotoxicity, or of the time in which that possible hazard really exists. Probably, the transmutation of minor actinides with fast fission processes is the most effective answer. This work, performed in SCK·CEN (Belgium) and DIMNP Pisa University, is focused on preliminary evaluation of industrial scale ADS (400 MWth, 2.5 mA) burning capability. An inert matrix fuel of minor actinides, 50% vol. MgO and 50% vol. $(Pu,Np,Am,Cm)O_{1.88}$, core content, with 150 GWd/ton discharge burn up, is used. The calculations were performed using ALEPH-1.1.2, MCNPX-2.5.0, and ORIGEN2.2. codes.

1. INTRODUCTION

This work aims to open a discussion about dedicated transmuters, connected to "*partitioning and transmutation (P&T) strategy*", namely, a subcritical reactor such as accelerator driven system (ADS).

In particular, in this paper, the preliminary burning capability efficiency of an industrial scale ADS (400 MWth, 2.5 mA) is evaluated [1].

Results obtained, with special regard to neutronic computational issues, have its basis in the contest of multipurpose hybrid research reactor for high-tech applications project (MYRRHA), see [2], experimental ADS design in development in Studiecentrum voor Kernenergie-Centre d'Étude de l'Energie Nucléaire, SCK·CEN, Belgium.

The starting point of this work is the present Belgian situation, where with seven PWR installed (5800 MWe), Belgian government decided (2003) to phase out nuclear energy production in the next 25 years, despite the opportunity of guarantying energy national independence and the engagement to respect the Kyoto agreement.

This was the incentive to propose an alternative scenario: the substitution of LWRs, the consequent installation of European pressurized reactor (EPR) in synergy with industrial scale ADS to support energy demand and, in the same time, to burn a consistent amount of HLW, could be a good connection to the foreseen possible fast reactors (Generation IV) [3].

Jointly, to reach an appropriated knowledge of transmutation strategy, the state of art of internal dosimetry in term of preliminary research was been done.

2. GENERAL ASPECTS

Global demand of energy increasing, oil and gas rise in prices, and significant climate changes are the majors issues of present century. To contain these phenomena, in order to obtain a sustainable energetic development, a critical and an integrate approach and the social acceptability of nuclear energy are nowadays necessary conditions.

Some strategies and technical solutions to minimize the quantities of long life radionuclides, as complementary approach to geological disposal of spent nuclear fuel (SNF), exist and are

(1) Pu recycling (MOX) in LWRs and FRs;

(2) MA and LLFP recycling in dedicated facilities as ADS;

(3) generation IV fast reactors able to burn the small, self-produced MAs and LLFPs amount.

Table 1: Fractions of Np, Am, and Cm upon the whole of minor actinides after a 1000 MWe PWR full life (58.5 years, under constant conditions).

Element	Weight fraction [%]
Np	0.315
Am	0.675
Cm	0.010

In the Integrated Project IP-EUROTRANS (EURATOM 6th Framework Programme—6th FP [4]), the preliminary concept of "European facility for industrial transmutation" (EFIT), can be found. His structure is based on the experiences gained during the 5th FP, where three experimental X-ADS designs (SCK·CEN, ANSALDO, and FRAMATOME) are defined.

3. COMPUTATIONAL TOOLS

To evaluate an industrial ADS burning efficiency, ALEPH-1.1.2 code [5, 6] was used. It is a Monte Carlo burnup code, under development at SCK·CEN in the framework of MYRRHA project, able to couple any version of MCNP [7] or MCNPX [8] with ORIGEN2.2 [9], for evolution calculation, using NJOY99.112 [10], for processing nuclear data in ENDF format (JEFF-3.1 [11]). MCNPX-2.5.0 and ORIGEN2.2 codes were used also separately from ALEPH-1.1.2, as explained further on.

4. NUCLEAR WASTE INVENTORY EVALUATION

Doel-3 (Belgium), a 1000 MWe PWR, is currently the reference reactor chosen in this work. Material discharged by this plant loaded by 4.3% enriched in U^{235} fuel was assumed to perform the calculations.

ALEPH-1.1.2 [5] code and JEFF-3.1 "evaluated data library" [11] were used to evaluate the inventory, considering a 50GWd/ton burnup and an irradiation cycle of 4.5 years.

Those specifications permitted to obtain weight fractions, in good agreement with literature values [12], referred to 1 ton of initial heavy metal (IHM), as 12.067 kg of Pu, 0.811 kg of Np, 0.282 kg of Am, and 0.128 Kg of Cm. The same values expressed as percentage are 1.06% wt. Pu/SNF, 0.072% wt. Np/SNF, 0.025% wt. Am/SNF, and 0.011% wt. Cm/SNF.

Noted that the quantities written above do not seem to play a significant role in health hazard and the choice to confine them in a geological disposal is the main solution adopted nowadays (once through scenario). But, also, nuclides half-life is related to social acceptability because these elements contribute to radiotoxicity for hundreds of thousands years.

Initial ADS fuel composition definition was performed on the basis of the waste inventory after the reactor full life, considering single batches decay: this value tries to represent a real PWR inventory composition.

So, we chose a 58.5 years of full life, in the context of PLEX strategy (Plan Lifetime EXtension). We simulated that any single batch undergoes 4.5 years of irradiation cycle (an ALEPH-1.1.2 code calculation), and we built a matrix in which every 1.5 years (length of internal fuel cycle) each batch is taken out from reactor and starts natural decay. In this way, hypothesizing constant working features during reactor full life, we estimated weight fractions upon the whole of minor actinides (Table 1).

5. RADIOTOXICITY ASPECTS

Radiotoxicity data from different sources imply often difficulties in understanding values and in comparing results among working groups; critical approach is then necessary to erase all uncertainties.

In this work, the radiotoxicity unit chosen is the "*Sv/GWe-y*," a suitablereference that allows to plot diagrams for a power plant radiotoxicity assessment independent from country law annual limit of intake for members of public.

Radiotoxicity coefficients data-bases referred to ICRP72 [13], that enable us to use an updated version of ORIGEN2.2 and ALEPH-1.1.2 codes, were created and considered as a further result of the described activity.

Also reference level (RL) needs a best estimated evaluation. We simulated (ORIGEN2.2 code calculation) the decay chains of natural uranium isotopic composition (g/cm^3) till to reach the secular equilibrium. We supposed that 1 ton of U 4.3%-enriched needed 8 tons of natural uranium, and we obtained a value of 3.07 10^6 Sv/GWe-y that corresponds to 1.61 10^5 Sv/ton IHM [1], in perfect agreement with international literature (NEA-OECD Report [12]).

6. THEORETICAL PARTITIONING & TRANSMUTATION

The assessment of theoretical partitioning and transmutation efficiency is the preliminary step to burning capability evaluation. In fact, it becomes a term of comparison.

Some definitions help to better understand this analysis:

(i) "*Transmutation efficiency*" defines the fraction of nuclides in SNF transformed in ones with shorter half-lives.

(ii) "*Transmutation cycle*" defines the irradiation period inside reactor; a decay period follows.

Within a single cycle, a transmutation efficiency close to unity is impossible to reach and it will be necessary an adequate study and an optimisation of subsequent cycles.

Acceptable time to reach reference level has been evaluated considering Pu a resource and not a waste and its partitioning efficiency. Separation of 99.9% wt. of Pu permits the achievement of RL in 2-3 $\times$ 10^4 years, considering the PUREX Process efficiency 99.9% wt [14].

After this evaluation, we assumed that a suitable time to reach RL, in order to consider the P&T acceptable, has to be 2, max 3, $\times 10^3$ years (Figure 1). In terms of MAs residual concentration left in wastes, the acceptable value obtained is about 27% wt. Theoretical transmutation efficiency, that is the reference value to tend forwards for ADS minor actinides burning capability, becomes equal to 73% wt. for all of minor actinides, in addition to 99.9% wt. Pu partitioning.

7. INDUSTRIAL ADS BURNING CAPABILITY

Fuel isotopic composition definition and core configuration are the first steps to calculate burning capability efficiency of an industrial scale ADS. We underline that this work is devoted to a preliminary evaluation and does not perform a full-scale calculation.

Industrial ADS structure (400 MWth, 2.5 mA, 600 MeV proton beam) modelled is pool-type reactor (Figure 2(a)), cooled by lead, with a diameter approximately 3 times bigger than height, characterized by $k_{\text{eff}} = 0.97$, 150 GWd/ton discharge burnup and loaded with CERCER fuel.

We considered an inert matrix fuel of minor actinides 50% vol. MgO and 50% vol. $(Pu,Np,Am,Cm)O_{1.88}$. In particular, the industrial size ADS studied consists in a hexagonal core and a central spallation loop separated from the core. Target, inside the spallation loop, consists of a windowless Pb free surface (design by MYRRHA project [2]). This layout allows a correct coupling with LINAR accelerator. The fuel assembly is hexagonal composed by a triangular lattice of cylindrical pins (Figures 2(b) and 2(c)). Basic data on the reactor facility, presented as EFIT development in 6th FP, have been furnished to the authors by MYRRHA project SCK·CEN research team (design data referred to March 2006).

Isotopic composition of ADS fuel was determined by successive steps.

(i) Fixing fractions of Np, Am, Cm on the whole of MA equal to the fraction calculated with accumulation during a PWR full life (Table 1). 20 years of natural decay, since the last batch has been extracted from reactor at the end of life, were taken.

(ii) Considering first generation plutonium in agreement with Belgian scenario chosen. This condition was dictated by Belgian government decisions: 2 reactors only, among 7, and just for a limited period of time (1995–2005), were loaded with MOX fuel.

(iii) Considering the nonstoichiometric ratio inside MA and Pu oxides equals to 1 : 1.88 instead of 1 : 2.

Some other steps became necessary to reach the suitable fuel composition, hypothesizing fuel density as 90% of theoretical density: at first, the calculation of ideal fuel solid solution within spots of $(Np,Am,Cm)O_{1.88}$; and second, the subsequent fuel and matrix ideal solid solution calculation.

At last, the quantity of first generation plutonium loaded in ADS fuel was minimized, in order to consider it a resource for future efficient development of fast reactors.

Different criticality calculations by MCNPX-2.5.0 [8] were then performed to respect the fixed $k_{\text{eff}} = 0.97$, where Pu concentration was a free parameter.

Taking into account these specifications, a composition of 37%at. Pu (atomic fraction) in 50% vol. fuel was obtained. Core was thought to be loaded with 324 homogeneous assemblies (Figure 3).

The originality of this work consists in considering the addition to Am and Cm of Np isotopes (Np-237 with $t_{1/2} = 2.144 \cdot 10^6$ y) inside ADS fuel.

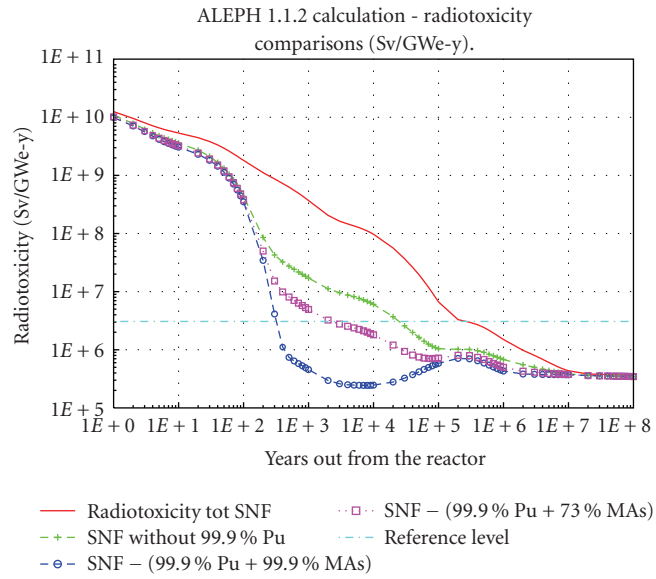

Figure 1: Theoretical transmutation efficiency of minor actinides: SNF (red line), SNF without 99.9% of Pu, and the reference levels are also represented.

Table 2: Burning capability for an industrial ADS: MA elements evolution during 3 transmutation cycles. (BOC: begin of cycle, EOC: end of cycle, and ETC: end transmutation cycle).

Element	Mass/initial mass			
	BOC1	EOC1	EOC2	ETC
Np	1.000	0.690	0.443	0.249
Am	1.000	0.693	0.454	0.271
Cm	1.000	2.478	3.202	3.592
Total MA	1.000	0.710	0.477	0.296

The simulated time period consists of 3 irradiation cycles (ALEPH-1.1.2 code calculation) of 2394 effective full power days (efpd) each, spaced out by decay periods of 3 years. 2394 efpd value is built on burn-up value, on thermal power of reactor and on quantity (ton) of initial heavy metal loaded (1). In particular, values chosen as reference are, respectively, 150 GWd/ton IHM, 400 MWth, and 6385 tonIHM, where

$$\text{efpd} = \frac{(\text{burn-up}) \cdot (\text{tonIHM})}{P\text{th}} \tag{1}$$

2394 efpd, equal to 6.6 years, was considered acceptable and in literature the same order of magnitude can be found, if referred to fast reactors [12].

This study was subsequently focused on the optimization of geometry and irradiation time parameters. Results obtained after nearly 2 months of parallel calculation in 4 CPU Dual Xeon 3 GHz are shown in Tables 2 and 3.

The brief overview of data written into tables makes possible to notice that, after the first cycle, transmutation efficiency (ϵ) is 29% wt. At the end of transmutation cycles (EOC3), if we consider total amount of MAs, a transmutation efficiency equal to 72% is achieved (Figure 4(a)).

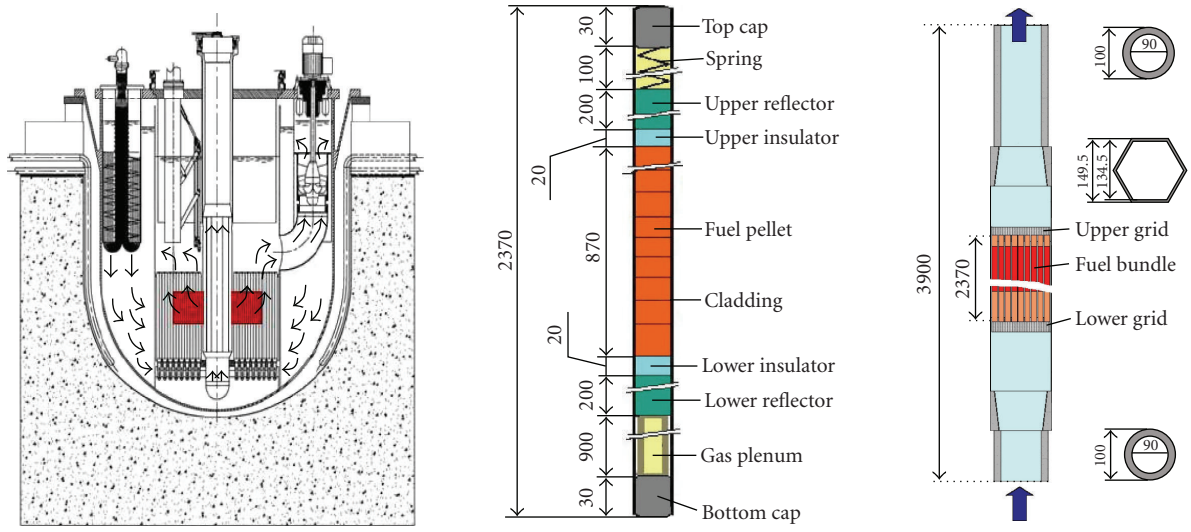

Figure 2: Conceptual configuration of EFIT (a). Simplified design of ADS fuel pin (b). Simplified design of ADS assembly (c).

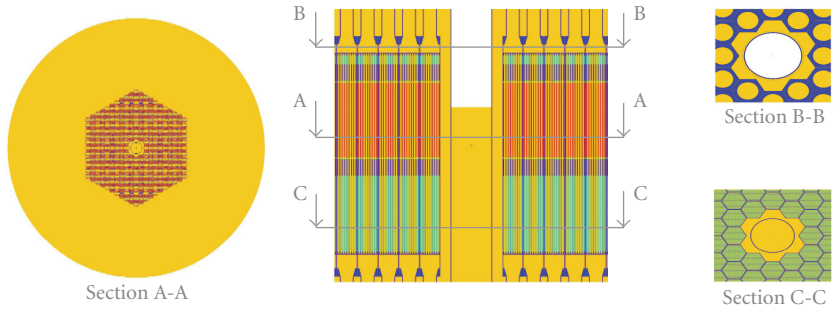

Figure 3: ADS MCNPX core model with 324 homogeneous fuel assemblies. Horizontal section (Section A-A). Particulars: assembly upper matching nozzle, $z = 92$ cm (Section B-B); fuel pin lower plenum in $z = -90$ cm (Section C-C).

Table 3: Burning capability for an industrial ADS: some isotopes evolution.

Isotope	Mass/initial mass			
	BOC1	EOC1	EOC2	ETC
Np237	1.000	0.690	0.443	0.249
Am241	1.000	0.645	0.391	0.211
Cm244	1.000	2.877	3.594	3.831

This value is in agreement with those values found with theoretical transmutation analysis described above.

In any case, the obtained values do not consider effective MAs separation efficiency (in particular the Cm one) necessary to prepare fuel for ADS.

Such preliminary remarks underline that a good result in transmutation and, then, in radiotoxicity reduction could be obtained just after more than 3 cycles inside ADS.

On the whole, total amount of MAs decreases during the burning processes; but some isotope content does not. Curium is an example. In order to describe better single elements amount during the transmutation period, that grow out of transmutation and decay chains contributions, mass to initial mass plus decay curves are presented (Figures 4(b), 4(c), 4(d)).

Curium inventory, accumulated in a single batch or during a LWR full life, is not too much (about 0.11 Kg Cm for each ton of SNF produced by LWR), but problems rise because of curium decay chain: it decays into Plutonium and increases total radiotoxicity.

About this last aspect, Cm-244 (Table 3), about 81.5% of total Cm with half-life of 18.1 years, increases at about 280% its initial quantity at the end of transmutation cycle (ETC). The Cm-244, in particular, decays into Pu-240, with half-life of 6563 years. So, Pu-240 inventory increases during transmutation (Figure 4(c)) and it brings a significant contribution to whole long time radiotoxicity.

8. CONCLUSIONS

Present LWRs and Generation IV fast reactors could be joined together by few ADS transmuters, built in limited number, in order to consider Pu a resource and not a waste.

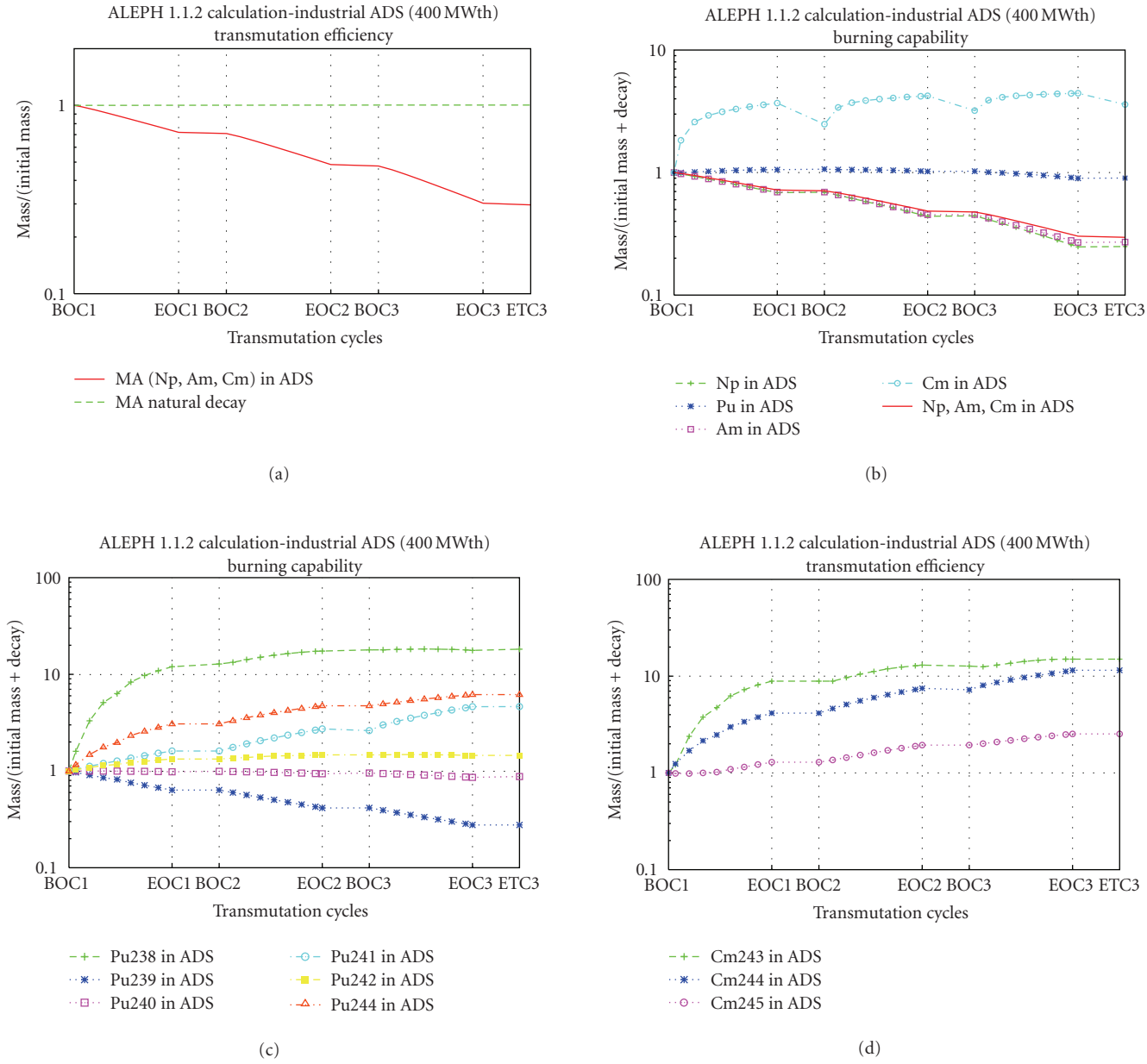

Figure 4: Industrial ADS burning capability during 3 transmutation cycles: mass to initial mass of total MAs evolution (a); evolution of single elements (b); evolution of Pu isotopes (c); evolution of Cm isotopes (d) [mass to initial mass plus decay].

In particular, the substitution of LWRs, the consequent installation of a new generation of passive safe reactors and industrial scale ADS to support energy demand and, in the same time, to burn a consistent amount of HLW, could be a good connection to Generation IV, in particular to fast reactors.

The definition of possible isotopic compositions of fuel and possible core configuration is the first step in order to calculate burning capability efficiency of an industrial scale ADS.

Moreover, creation of radiotossicity coefficients databases improves a more efficient and flexible use of ALEPH-1.1.2 and ORIGEN2.2 codes.

NOTATIONS

Efpd: Effective full power days
BU: Burn-up
Ton: Fuel tons initially loaded in ADS
*P*th: Thermal power
E: Transmutation efficiency
K_{eff}: ADS multiplication factor

ABBREVIATIONS

ADS: Accelerator driven system
P&T: Partitioning & transmutation

PWR: Pressurized water reactor
LWR: Light water reactor
EPR: European pressurized reactor
HLW: High level waste
SNF: Spent nuclear fuel
MOX: Mixed oxide
FR: Fast reactor
MA: Minor actinides
LLFP: Long live fission product
EFIT: European facility for industrial transmutation
ENDF: Evaluated nuclear data file
IHM Initial heavy metal
PLEX: Plan life-time extension
ICRP: International commission on radiological protection
RL: Reference level
PUREX: Pu and U recovering by extraction process
LINAC: LINear accelerator
BOC: Begin of cycle
EOC: End of cycle
ETC: End transmutation cycle
MYRRHA: Multipurpose hybrid research Rreactor for high-tech applications
CERCER: Ceramic-ceramic fuel

REFERENCES

[1] B. Calgaro and B. Vezzoni, *Contribution to long term minor actinide management through transmutation in accelerator driven systems (ADS)*, M.S. thesis, University of Pisa, Pisa, Italy, 2006.
[2] H. Aït Abderrahim, et al., "MYRRHA pre-design file—draft 2," Tech. Rep. R-4234, Studiecentrum voor Kernenergie, Centre d'Etude de l'Energie Nucléaire (SCK·CEN), Brussels, Belgium, 2005.
[3] B. Verboomen, et al., "Working party on scientific of the fuel cycle the Belgian implementation scenario," Studiecentrum voor Kernenergie, Centre d'Etude de l'Energie Nucléaire (SCK·CEN), Belgium, 2005.
[4] 6thFP EURATOM, "Integrated Project EUROpean Research Programme for the TRANSmutation of High Level Nuclear Waste in an Accelerator Driven System," 2005.
[5] W. Haeck and B. Verboomen, "ALEPH-1.1.2, a Monte Carlo burn-up code, SCK-CEN-BLG-1003 Rev.0," Studiecentrum voor Kernenergie, Centre d'Etude de l'Energie Nucléaire (SCK·CEN), Belgium, January 2006.
[6] W. Haeck and B. Verboomen, "An optimum approach to Monte Carlo burn-up," *Nuclear Science and Engineering*, vol. 156, no. 2, pp. 180–196, 2007.
[7] J. F. Briesmeister, "MCNP—a general Monte Carlo N-particle transport code, version 4C, LA-13709-M," Los Alamos National Laboratory, USA, 2000.
[8] D. B. Pelowitz, Ed., "MCNPX user's manual, version 2.5.0, LA-CP-05-0369," Los Alamos National Laboratory, USA, April 2005.
[9] A. G. Croff, "A user's manual for the ORIGEN2 computer code, ORNL/TM-7175," Oak Ridge National Laboratory, USA, July 1980.
[10] NEA, "Joint Evaluated Fission and Fusion File (JEFF-3.1)," OECD, Paris, France, 2005.
[11] R. E. MacFarlane and D. W. Muir, "The NJOY nuclear data processing system version 91, LA-12470-M," Los Alamos National Laboratory, USA, 1994.
[12] NEA, "Physics and safety of transmutation systems: a status report," Tech. Rep. 6090, OECD, Paris, France, 2006.
[13] ICRP72, "Recommendation of the International Commission on Radiological Protection, ICRP Publication 72," Pergamon Press, Oxford, UK, 1996.
[14] NEA, "Advanced nuclear fuel cycles and radioactive waste management," Tech. Rep. 5990, OECD, Paris, France, 2006.