N-15 Requirement for 2nd Stratum ADS Nitride Fuels

J. Wallenius
Department of Nuclear and Reactor Physics
Royal Institute of Technology
Stockholm Centre for Physics, Astronomy and Biotechnology
S-106 91 Stockholm

S. Pillon
DEN/DEC
CEA Cadarache Bât 315
F-13108 Saint-Paul-lez-Durance

Abstract

The application of nitride fuels in accelerator driven systems dedicated to waste transmutation requires use of N-15 enriched nitrogen to suppress C-14 production due to (n,p) reactions on N-14. With C-14 emission rates of the oxide reprocessing plants as reference, we find that 99% N-15 enrichment is sufficient when using nitride fuels for minor actinide transmutation in 2nd stratum accelerator driven systems. Projected cost estimations make recycling of N-15 in the fabrication stage mandatory, while recovery of N-15 from reprocessing does not appear to be necessary.

INTRODUCTION

If one by partitioning and transmutation wishes to reduce the long term radiotoxic inventory directed to geological repositories by a factor of 100 or more, recycling of americium and curium becomes mandatory [1]. As the potential for homogeneous recycling of higher actinides in critical reactors is limited [2-4], the Double Strata fuel cycle was suggested by JAERI [5]. In this concept, minor actinide waste from commercial reactors are recycled in dedicated reactors that operate on fuel free from U-238. Due to an decrease in Doppler feedback, an increase in void worth and a very small fraction of delayed neutrons, it is assumed that such MA-burners should operate in sub-critical mode in order to meet safety requirements [2,6].

The composition of the fuel to be used in Accelerator Driven Systems (ADS) dedicated to waste transmutation has yet to be determined. Possible choices are oxides, nitrides and metals, each one featuring advantages and drawbacks. In the European research program on partitioning and transmutation, oxides are regarded as the preferred option, due to fewer problems related to fabrication and the huge accumulated experience of oxide fuels taken to high burnup in various fast reactor programs.

However, since plutonium oxide is not soluble in nitric acid without the use of special catalysts, standard PUREX reprocessing is difficult to apply to uranium free oxide fuels. Therefore pyrometallurgical methods have to be developed for this purpose. Plutonium nitride on the other hand is soluble in nitric acid, and nitride fuels hence appear to offer a larger flexibility in the choice of reprocessing methods.

In the European Union CONFIRM program [7], uranium free nitride fuels are explored as a backup option for application in Accelerator Driven Systems. An important issue to clarify in the present context, is the enrichment level of N-15 required to avoid excessive production of C-14 due to (n,p) reactions on N-14. In the present paper, this problem is discussed taken into account the specific features of the Double Strata fuel cycle. First, we calculate the C-14 production in an ADS typical for the 2nd stratum as function of N-15 enrichment. Then we make an estimation of the total C-14 production arising from both first and second stratum of a full P&T scenario. Comparing the production rates, we arrive at a value for the N-15 enrichment level that will ensure an acceptable increase in C-14 emission from the reprocessing plant due to the use of nitride fuel in the 2nd stratum.

CARBON-14

C-14 is one of the main contributors to the exposure of radio-toxicity in the vicinity of reprocessing plants, as carbon is typically released in the off-stream gas. C-14 is mainly produced in (n,p) reactions on N-14, that is present as an impurity in oxide fuel as well as in cladding and structural material. According to a study made by BNFL, 4.7% of the dose to the group of

individuals most prone to inhalation exposure from THORP in Sellafield is due to C-14 [8].

One may thus argue that if nitride fuels are to be used anywhere in the nuclear fuel cycle, the resulting additional production of C-14 should not lead to a significant increase in dose committments. There are several ways to limit such releases:

1) Use of nitride fuels only in a limited part of the fuel cycle
2) Use of N-15 enriched nitrogen for the fabrication of nitride fuels
3) Implementation of carbon trapping in the off-stream following reprocessing

The present investigation concerns a combination of the first and second of the above.

THE DOUBLE STRATA SCENARIO

For the present analysis, the Double Strata scenario [5, 6] was adopted. In the particular implementation [9], plutonium from spent UOX LWR fuel is recycled once in LWRs and then multi-recycled in fast neutron reactors of CAPRA type. All minor actinides are directed to accelerator driven systems, where they are multirecycled together with the low quality Pu present in ADS discharges. A nuclear park producing 1000 TWhe was assumed. This corresponds to 140 GWe installed power, assuming an average availability of 80%. According to the NEA study, 6.0 percent of the park power is produced by accelerator driven systems, 19.5 percent by the CAPRA reactors, and 74.5 percent by light water reactors operating on UOX and MOX fuel [9]. The minor actinide flow rate into the 2nd stratum is 6.6 kg/TWhe, or 6.6 tons per year for the park considered. The ADS is assumed to operate on uranium free nitride fuel.

ADS CORE SIMULATIONS

In order to make an accurate calculation of the C-14 production rate in the ADS, a fully three-dimensional (pin by pin) model of a sub-critical core similar to the one proposed by JAERI was made [10]. The Monte Carlo codes MCNPX was used for simulation of proton and neutron transport in the core [11]. The characteristics of the core setup are displayed in Table 1. Note in particular the presence of zirconium nitride in the fuel. While PuN is unstable towards dissociation at temperatures below its melting point, calculations based on measured solidus and liquidus temperatures of $(U_{0.8},Zr_{0.2})N$ indicate that (Pu,Zr)N should remain stable up to melting [7]. The increase in nitrogen inventory due to the presence of ZrN will however lead to additional production of C-14. In the present study the relative fraction of ZrN in the fuel was determined by the following procedure:

The number of fuel pins was considered to be fixed by setting the core power to 820 MW and adopting an average linear rating of 30 kW/m. Postulating 10% heavy atom burnup per year and a TRU fission rate of 240 kg per year then yields a TRU inventory of 2400 kg, corresponding to 90g/pin. The Pu to MA ratio at BOL was considered to be fixed by burnup swing minimisation to Pu:MA = 40:60 [10]. The pin pitch of 1.45 PD was determined by a imposing a coolant temperature rise less than 100 K over the core. Zirconium nitride was finally added to the fuel until a k-eigenvalue equal to 0.95 was obtained. This condition was typically fullfilled for equal molar fractions of TRU and Zr.

Burnup was calculated using MCB, a newly developed Monte Carlo Burnup code based on MCNP [12] and developed at KTH. Continuous cross section libraries for more than 300 nuclides and an energy dependent fission product yield library was used for calculating transmutation trajectories. Cross sections, fluxes and other core properties were recalculated with a time step of 100 days for a core power of 820MWth.

The N-15 enrichment level of the 300 kg nitrogen present in the core was set to be 99%. Hence uncertainties in C-14 production rates due to self-shielding effects at high enrichment levels could be avoided. The core averaged cross section for the $^{14}N(n,p)$ reaction equaled 15 mb. The resulting production rate of C-14 in the fuel was 6.7 grams per 300 equivalent full power days. Adopting an availability factor of 0.8, 6.7 grams C-14 equals the annual production of the core.

Noting that cladding and construction material are significant contributors to the C-14 production in LWRs (up to 60%), this source of C-14 should also be considered. Assuming an N-14 contamination of 250 ppm weight in the clad, 0.8 grams C-14 had been produced in the cladding after 300 days. As long as the clad itself is not dissolved, it will however not contribute to the atmospheric release of C-14.

Table 1: *Characteristics of the ADS used in the analysis*

Core Power	820 MWth
Linear rating (average)	30 kW/m
Spallation target	PbBi – 19 cm radius
Sub-criticality at BOL	0.95
Coolant	Sodium
Fuel	$(Pu_{0.2},MA_{0.3},Zr_{0.5})^{15}N$
Clad outer/inner diameter	5.9/5.0 mm
Pin Pitch	1.45 PD
Burnup	20% h.a. / 600 EPFD

NITRIDES VERSUS OXIDES

With 6.6 tons of Minor Actinides to be managed by accelerator driven systems (see Figure 1), 28 of the above ADS cores would be present in the 1000 TWhe park. LWR plutonium is assumed to be needed only for startup. The annual C-14 production due to the use of nitride fuel with 99% N-15 enrichment then becomes 185 grams.

The release of C-14 from reprocessing of oxide fuels being about 3 g/(GWyear) [13], the contribution of the 1st stratum light water reactors amounts to 300 grams annually. The Pu managing CAPRA reactors provide another 70 grams, yielding a total C-14 release from oxide fuels equal to 370 grams.

Introduction of nitride fuels for MA transmutation in ADS thus increases releases of C-14 into the atmosphere by about 50%, for an N-15 enrichment level of 99%. As C-14 from oxide reprocessing provides less than five percent of the total inhalated dose to high risk individuals, one may conclude that this enrichment level is sufficient to avoid a significant increase in dose committments.

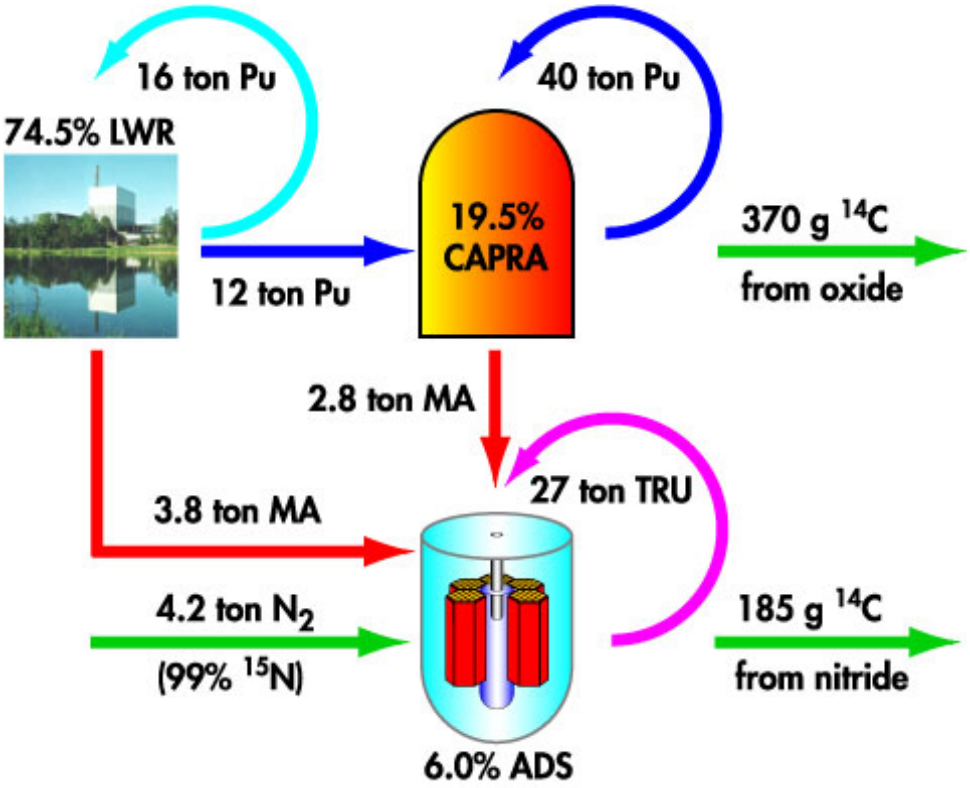

Figure 1: *Mass flows and C-14 production in the 1000 TWhe nuclear park analysed in the present paper. LWRs and Fast Neutron CAPRA reactors use oxide fuels. The ADS is assumed to operate on nitride fuel, where the nitrogen is enriched in N-15.*

NITRIDE FABRICATION

Pending calculations of cladding damage, we set the fuel residence time to two years, yielding a final average burnup of 20 percent heavy atoms. Consequently, 33.2 ton of heavy metal needs to be fabricated into ADS fuel each year, demanding 4.2 tons of nitrogen. In existing fabrication lines, an open gas cycle is used for converting oxides or metals into nitrides, and typically about 99% of the nitrogen supplied is lost. A closed gas cycle would therefore have to be implemented. Estimations made at CEA show that about 200 percent of excess gas inventory could be sufficient in the fabrication of a given amount of nitride. Being a one time expense (depending on frequency of accidental gas lossses), one could expect a cost penalty of less than 20 percent relating to the excess inventory.

N-15 ENRICHMENT

N-15 is today used as a tracer in agronomic and organic industry. The world wide market is about 20 to 40 kilograms annually, and the current cost is about 80 euro/g. The supply of several tons per year to the 60

TWhe ADS park, would therefore demand an increase in production capacity by a factor of 100. Accordingly, the specific production cost can be expected to decrease. Estimations made at CEA point towards a price of approximately 10 euros per gram, depending on the selected method of fabrication. The total cost for the N-15 supply would then approach 50 million euro annually. This number should be compared to the total production cost for 1000 Twhe in the nuclear park here studied.

The cost of electricity from LWRs operating on UOX fuel is today well known, and a recent study from the university of Lappenranta predicts a cost of 22 keuro/GWhe for a planned reactor block (1.0 – 1.5 Gwe) built at an existing reactor site. Currently the price of LWR MOX is four times that of UOX. Fuel representing 12% of the total cost for the future Finnish reactor, the price for electricity from light water reactors operating on a mix of UOX and MOX can be calculated to 23 Meuro/TWhe.

The cost of fast reactor electricity is less well established. Figures from different studies vary from the 30 000 euro/GWhe for the lead cooled BREST-1200 reactor to 40 000 euro/GWhe for BN-800. The cost increment for ADS electricity is obviously based on theoretical estimations only, but a recent NEA study arrived at a preliminar figure of 60 000 euro/GWhe for a nitride fueled ADS [14]. This is in reasonable agreement with a study of relative fuel cycle cost made at JAERI. We thus arrive at an average cost for electricity produced in the present park equal to 27 Meuro/TWh, with the uncertainty estimated to be about 5 percent. The total cost of electricity (excluding N-15 enrichment) then becomes 26 – 28 Geuro per year. Comparing with the above estimation for the N-15 enrichment cost (50 Meuro/year) one finds that its relative contribution to the total cost is of the order of 0.2 percent, which should be acceptable.

CONCLUSIONS

Under the assumptions made in the present study of the Double Strata fuel cycle, 6.0% of the total nuclear park power is produced by minor actinide transmutation in ADSs operating on nitride fuels. It has been found that for an N-15 enrichment level of 99%, the C-14 production in ADS fuel is half of that produced in the oxide fuels used in the first stratum. Noting that presently C-14 constitutes 5% of the dose to the group most exposed to inhalation doses from THORP , it is our opinion that the increase in dose committments is acceptable. C-14 releases from reprocessing of ADS nitride fuels would however become a major contributor to inhalated doses at N-15 enrichment levels less than 90% (Provided that carbon trapping is not implemented). Hence we conclude that the required enrichment level for nitrogen to be used for nitride fuel fabrication is in the range of 98-99 percent, depending on the actual fraction of nitrogen containing inert matrix in the fuel. Use of higher enrichemt levels would not be meaningful as long as carbon trapping is not implemented for the reprocessing of oxide fuels.

The cost for the use of N-15 could be estimated to 50 Meuro/year, corresponding to 0.2% of the total cost for the 1000 Twhe park. In our opinion this is an acceptable penalty. The biggest uncertainties in the cost analysis are:

a) The fraction of inert matrix nitrogen to be supplied, which could increase the N-15 cost by up to a factor of two;
b) The frequency of accidental loss of gas in the fabrication facility, which would increase the cost by a factor of three, if the full gas inventory would have to be replaced annually.

Implementation of N-15 recovery from reprocessing could further decrease the penalty, but does not appear to be required. On the other hand, the use of a closed gas cycle in the fabrication stage is clearly mandatory.

ACKNOWLEDGEMENTS

This work was made as part of the CONFIRM project, funded by the European Commission.

REFERENCES

1) M. Delpech et al, The Am and Cm transmutation, physics and feasibility. Proc. Int Conf. Future Nuclear Systems, GLOBAL 99. ANS 1999.

2) D.G. Foster et al, Review of PNL study on transmutation processing of high level waste. LA-UR-74-74, LANL 1974.

3) S.L. Beaman, Actinide Recycle in LMFBRs as a waste managemnet alternative. Proc. 1st Int. Conf. Nuclear Waste Transmutation, page 61. University of Texas (1980).

4) J. Tommasi et al, Long lived waste transmutation in reactors, Nucl. Tech. 111 (1995) 133.

5) H. Murata and T. Mukaiyama, Fission reactor studies in view of reactor waste programs. Atomenergie-Kerntechnik 45 (1984) 23.

6) M. Salvatores et al, Long-Lived Radioactive waste transmutation and the role of accelerator driven (hybrid) systems. Nucl. Inst. Meth. A, 414 (1997) 5.

7) J. Wallenius et al, The European Union CONFIRM project, these proceedings.

8) S. Beaty, The THORP project – an overview. Energy and environment 6 (1995) 383.

9) P. Wydler, P&T studies of the OECD/NEA, Fifth framework program meeting on P&T and ADS activities, Paris 2000.

10) T. Takizuka et al, Studies on accelerator driven transmutation systems. In Fifth international infomation exchange meeting on actinide and fission product partitioning and transmutation, page 383. EUR-18898 EN, OECD/NEA, 1998.

11) L. Waters, editor, MCNPX user's manual, version 2.1.5 TPO-E83-G-UG-X-00001, LANL 2000.

12) J.F. Briesmeister, editor, MCNP – A general Monte Carlo N-Particle transport code, version 4C, LA-13709-M, LANL 2000.

13) W.R.A. Gossens, C.G. Eichholz and DW. Tedder, eds, Treatments of gaseous effluents at nuclear facilities, Radioactive waste management handbook vol 2 (1991).

14) D. Westlen et al, A cost benefit analysis of an accelerator driven system, these proceedings.