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## Plutonium and Minor Actinide Management in Thermal High-Temperature Gas-Cooled Reactors Publishable Final Activity Report

Author(s):

J.C. Kuijper, J. Somers, L. Van Den Durpel, V. Chauvet, N. Cerullo, J. Cetnar, T. Abram, K. Bakker, E. Bomboni, W. Bernnat, J.G. Domanska, E. Girardi, J.B.M. de Haas, K. Hesketh, J.P. Hiernaut, K. Hossain, J. Jonnet, Y. Kim, J.L. Kloosterman, M. Kopec, J. Murgatroyd, D. Millington, D. Lecarpentier, G. Lomonaco, D. McEachern, A. Meier, M. Mignanelli, H. Nabielek, J. Oppe, B.Y. Petrov, C. Pohl, H.J. Rütten, S. Schihab, G. Toury, C. Trakas, F. Venneri, K. Verfondern, H. Werner<sup>†</sup>, T. Wiss, J. Zakova

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**PUMA (Plutonium & Minor Actinide Management in Thermal High Temperature Reactors)**







EC Scientific Officer: **Panagiotis MANOLATOS**

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to: P. Manolatos (EC Scientific Officer, Brussels) Partners Euratom project "PUMA" (contract no. FP6-036457)

from: : J.C. Kuijper (PUMA Coordinator) NRG/Petten/Fermi-building

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## Summary

The **PUMA** project -the acronym stands for "Plutonium and Minor Actinide Management in Thermal High-Temperature Gas-Cooled Reactors"- was a Specific Targeted Research Project (STREP) within the Euratom 6th Framework (EU FP6). The PUMA project ran from September 1, 2006, until August 31, 2009, and was executed by a consortium of 14 European partner organisations and one from the USA.

This report serves 2 purposes. It is both the "**Publishable Final Activity Report**" and the "**Final (Summary) Report**", describing, per Work Package, the specific **objectives, research activities, main conclusions, recommendations** and **supporting documents**.

PUMA's main objective was to investigate the possibilities for the utilisation and transmutation of plutonium and especially minor actinides in contemporary and future (high temperature) gas-cooled reactor designs, which are promising tools for improving the sustainability of the nuclear fuel cycle. This contributes to the reduction of Pu and MA stockpiles, and also to the development of safe and sustainable reactors for CO<sub>2</sub>-free energy generation.

The PUMA project has assessed the impact of the introduction of Pu/MA-burning HTRs at three levels: fuel and fuel performance (modelling), reactor (transmutation performance and safety) and reactor/fuel cycle facility park.

Earlier projects already indicated favourable characteristics of HTRs with respect to Pu burning. So, core physics of Pu/MA fuel cycles for HTRs has been investigated to study the CP fuel and reactor characteristics and to assure nuclear stability of a Pu/MA HTR core, under both normal and abnormal operating conditions. The starting point of this investigation comprised the two main contemporary HTR designs, viz. the pebble-bed type HTR, represented by the South-African PBMR, and hexagonal block type HTR, represented by the GT-MHR. The results (once again) demonstrate the flexibility of the contemporary (and near future) HTR designs and their ability to accept a variety of Pu- and Pu/MA-based fuels (possibly in combination with thorium), and to obtain a significant reduction of the Pu-respectively Pu/MA content, while maintaining, to a large extent, the well-known standard (U-fuelled) HTR safety characteristics. However, this will require some changes in the reactor design. Studies have furthermore shown that fuel with a "diluted" kernel ("inert-matrix") improves the transmutation performance of the reactor.

A study on proliferation resistance, taking into account several possible proliferation pathways, highlights that a prismatic (V)HTR core would be amenable to conventional safeguards accounting and verification procedures, with fuel blocks accounted for individually in the same way as LW fuel assemblies. However, a modified approach would be needed in pebble bed cores because of the impracticability of accounting for individual fuel spheres.

When dealing with minor actinide bearing fuel helium generation is an important issue. Experiments have shown that He will be released from the kernel, but not from fresh kernels. Indeed, fresh fuel has shown a remarkable stability – up to 2500 °C. For modelling purposes, 100% release of helium from the kernel is justified. The diluted kernel concept was first invoked by Belgonucleaire brings many benefits. The fuel modelling studies have clearly indicated the advantages that can be gained by dilution. Essentially, for a given buffer layer thickness, more volume is available to accommodate the CO and He released. Chemical thermodynamic models have been deployed to design a kernel that will show limited CO production. The most important point here is that substoichiometric Pu or Am oxides are essential. Further improvement can be achieved by chemical buffering of the fuel by the addition of Ce sesquioxide, which takes up oxygen to form the dioxide. Ultimately any coated particle design must be validated in an irradiation test. Though not possible to perform an irradiation programme in the PUMA project, the feasibility of such a programme has been demonstrated, and the initial data needed to launch such a test has been generated.

Pu/MA transmuters are envisaged to operate in a global system of various reactor systems and fuel cycle facilities. Fuel cycle studies have been performed to study the symbiosis to other reactor types/systems, and to quantify waste streams and radiotoxic inventories. This includes studies of symbiosis of HTR, Light Water Reactor (LWR) and Fast Reactor (FR) systems, as well as the assessment of the technical, economic, environmental and socio-political impact. It is e.g. shown that a Pu/MA-loaded HTR may have a considerable, positive impact on the reduction of the amount of TRU in disposed spent fuel and high level waste.

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## The PUMA Consortium

The PUMA group: 16 partners from 8 Member States, the EU and the USA

1. NRG – Nuclear Research & consultancy Group	The Netherlands
2. AGH – Univ. of Science and Technology of Cracow	Poland
3. BN – Belgonucleaire	Belgium
4. CIRTEN	Italy
5. EDF – Electricité de France	France
6. GA – General Atomics	USA
7. USTUTT – University of Stuttgart	Germany
8. JRC – Joint Research Centre	EU
9. KTH – Royal Institute of Technology	Sweden
10. LISTO evba	Belgium
11. NEXIA – Nexia Solutions Ltd.	United Kingdom
12. NNC – National Nuclear Corporation	United Kingdom
13. ( <i>vacant</i> )	
14. LGI – LaGrange Innovation	France
15. TUD – Delft University of Technology	The Netherlands
16. FZJ – Forschungszentrum Jülich	Germany
17. ANP – Areva NP SAS	France

### **Contact details of the PUMA Coordinator**

Dr. Jim C. KUIJPER  
NRG  
Westerduinweg 3  
PO Box 25  
Petten, The Netherlands  
Phone: + 31 224 564506  
Fax: + 31 224 568490  
Email: [kuijper@nrg.eu](mailto:kuijper@nrg.eu)

### **Web**

Project public website: <http://www.puma-project.eu>  
Project private website: <https://members.puma-project.eu>





# 1 Project execution

## 1.1 Project objectives and relation to the state-of-the-art

*This section presents the objectives of the PUMA project at the start of the project, 1. September 2006.*

The sustainability of the nuclear fuel cycle and the reduction of plutonium (Pu) and Minor Actinides (MA) stockpiles are key issues in the definition of the future nuclear energy mix in Europe. The High Temperature gas-cooled Reactor (HTR) can fulfil a very useful niche for the purposes of Pu and MA incineration due to its unique and unsurpassed safety features, as well as to the attractive incentives offered by the nature of the coated particle fuel.

No European reactor of this type is currently available, but there have been two test reactors (DRAGON, AVR) and one 300 MW<sub>el</sub> demonstrator (THTR) successfully operated creating the substantial know-how, which is still available. Considerable interest in the HTR is growing internationally including projects to construct modular HTR prototype reactors in China and in South Africa. In Europe, further development of HTR has been undertaken within the “Reactor for Process Heat, Hydrogen and Electricity (RAPHAEL<sup>1</sup>)” Integrated Project of the 6<sup>th</sup> Framework Programme.

Apart from the inherent safety features offered by this reactor type, the nature of the coated particle (CP) fuel offers a number of attractive incentives. In particular, it can withstand burn-ups far beyond that in either LWR or FR systems. Demonstrations of Pu-burning as high as 75% fissions in initial metal atoms (FIMA) have been achieved in former tests. In addition, the coated particle itself offers significantly improved proliferation resistance, and finally with a correct choice of the kernel composition, it can be a very effective support for direct geological disposal of the spent fuel without significant Instant Release Fraction.

Complementary with other initiatives, **the objective of the PUMA project was, and is, to provide key results for the utilisation and transmutation of plutonium and minor actinides in HTRs.**

A number of important issues concerning the use of Pu and MA in gas-cooled reactors have already been studied in other projects. These, and other, earlier projects show favourable characteristics of HTRs with respect to Pu burning and subsequent disposal of HTR spent fuel e.g. with regard to leach resistance and lower decay heat. It has to be mentioned that the CP retains not only residual MA within the final repository but also the long-lived fission products, which are much more soluble and mobile than MA. However, further steps are required to demonstrate the potential of thermal HTRs as Pu/MA transmuters based on realistic/feasible designs of CP Pu/MA fuel. Therefore, the overall objective of the PUMA project is to provide **additional key elements** for the utilisation and transmutation of plutonium and minor actinides in contemporary and future (high temperature) gas-cooled reactor design. High temperature reactors are promising systems for improving the sustainability of the nuclear fuel cycle, for reducing Pu and MA stockpiles and for developing safe and sustainable reactors for CO<sub>2</sub>-free energy generation.

For that reason, core physics of Pu/MA fuel cycles for HTRs are investigated to optimise the CP fuel and reactor characteristics and to ensure nuclear stability of a Pu/MA HTR core.

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<sup>1</sup> [www.raphael-project.org](http://www.raphael-project.org)

It is also in the scope of PUMA to optimise the present Pu CP design and to explore the feasibility of MA fuel. New CPs will be designed that can withstand very high burn-ups and are well adapted for disposal after irradiation. The project benefits greatly from access to past knowledge from Belgonucleaire's Pu HTR fuel irradiation tests of the 1970's, and also secures access to materials made at that time.

(Very) High Temperature Reactor V/HTR Pu/MA transmuters are envisaged to operate in a global system of various reactor systems and fuel cycle facilities. Fuel cycle studies are also included in PUMA, to study the symbiosis between LWR, GCFR and ADS, and to quantify waste streams and radiotoxic inventories. The technical, economic, environmental and socio-political impact is assessed as well.

*The following sections present a description of the main **objectives, activities, conclusions and recommendations** of the technical Work Packages (1-3) and the associated tasks. As such this report serves two purposes. It is both the "Publishable Final Activity Report" and a "Final (Summary) Report" of the PUMA project.*

## 1.2 Work Package 1

### 1.2.1 Introduction

#### 1.2.1.1 Reminder of objectives of Work Package 1 (WP1)

The first PUMA Work Package was concerned with the core physics, including transient behaviour, of Pu/MA loaded HTRs. At the start of the project the main objectives of this Work Package were:

- Demonstration of the full potential of contemporary and (near) future HTR designs to utilise/transmute Pu and minor actinide fuel within the constraints of safe operation, and based upon realistic assumptions concerning the fuel composition.
- To identify necessary additional qualification of the tools employed for the assessment of Pu/MA-loaded HTR systems, and identify opportunities to obtain experimental data on which such additional qualification can be based. Such opportunities will e.g. be sought in cooperation with other EU FP6 projects as well as with external organisations, such as the American GT-MHR programme (through partner GA), as well as British (through partners NEXIA and NNC) and Russian institutes (specifically the ASTRA critical facility at the Kurchatov Institute, Moscow, through ISTC and IAEA/INPRO), and the OECD/NEA. The latter is to ensure that descriptions, on which additional code qualification exercises are to be based, are well-defined and complete. For the code qualification (benchmark) exercises the intention is to adhere to OECD/NEA IRPhE requirements.

However, the main task within this particular Work Package is concerned with the assessment of several HTR system/fuel/fuel cycle combinations. Based on results obtained in particular in EU FP5 projects HTR-N/-N1, the core physics work in PUMA was envisaged to establish optimised Pu/MA transmutation characteristics in HTRs. Transient analyses demonstrate the nuclear stability and safety of the optimised reactor/fuel designs. The results also comprise measures on other performance and safety-related parameters, such as the fast fluence in the CP coatings and the **helium production in HTR coated particles** (D131), as well as an **assessment of proliferation resistance** (D132) Reference systems for these studies are contemporary representatives of the two main HTR designs, viz. the **PBMR-400** for the pebble-bed (continuous reload) type, and the **GT-MHR** (or derivative) for the prismatic block type, loaded with Pu-based (U-free) fuel (D121). Studies include:

- Pu/MA deep burn in pebble-bed V/HTR. This also includes investigations on the utilisation of IMF, such as  $(Zr,Y,Am)O_2/(Zr,Y,Ce)O_{2-x}$  or  $(CeAm)O_{2-x}$  (D122);
- Th/Pu fuel cycle in pebble-bed V/HTR (D123);
- Pu/MA deep burn in prismatic V/HTR (D124);
- Th/Pu fuel cycle in prismatic V/HTR (D125);
- Reactivity transients of Pu/MA fuelled V/HTR (D126);
- Integrated LWR-HTR-GCFR symbiotic fuel cycles (D127).

#### 1.2.1.2 Contributions to WP1

The participants in this Work Package were NRG (WPL), AGH, BN, CIRTEN, EDF, FZJ, GA, KTH, NNL and USTUTT. During the project BN ceased to operate, but their contribution (to

D131; in support document S133) was at least partly achieved prior to the cessation. The remaining part was taken up by AGH. The WP1 deliverables (*Dnnn*) and support documents (*Snnn*) are listed below. Their output, conclusions and recommendations are summarised in this report. Further references are given in each of the respective sections of this report.

No.	Title	Responsible
D102	Summary report on Work Package 1	NRG (Kuijper)
D111	Report on required additional qualification of analysis tools, possibilities for obtaining relevant experimental data & additional qualification exercise	KTH (Zakova)
D112	Report on additional code qualification opportunities	NRG (Petrov, Oppe)
D121	Report on selected reference HTR designs and fuel cycle data	NRG (Kuijper)
D122	Report on assessment of “deep burn” Pu/MA utilisation in pebble bed [V]HTR	NRG (Petrov)
D123	Report on assessment of Th/Pu fuel cycle in pebble bed [V]HTR	FZJ (Pohl)
D124	Report on assessment of “deep burn” Pu/MA utilisation in prismatic [V]HTR	AGH (Cetnar)
D125	Report on assessment of Th/Pu fuel cycle in prismatic [V]HTR	EDF (Girardi)
D126	Report on reactivity transients of Pu/MA fuelled [V]HTRs	USTUTT (Bernnat, Meier)
D127	Report on the assessment of LWR-HTR-GCFR integrated fuel cycles	CIRTEN (Lomonaco)
D131	Report on the calculation analysis of He production in HTR coated particle fuel	AGH (Cetnar)
D132	Report on the assessment of proliferation resistance	NNL (Hesketh)
S128	PUMA Pebble-Bed HTR Reactor and Fuel Data for Use in the DANESS Code	NRG (Petrov, Kuijper)
S133	Prediction of helium production in coated particles	BN (Shihab, Toury)

### 1.2.1.3 Pu/MA HTR reference systems and fuel compositions (D121)

#### *Authors*

J. Cetnar (AGH),  
J.B.M. de Haas (NRG)  
J.C. Kuijper (NRG)  
D. McEachern (GA)  
J. Oppe (NRG)  
F. Venneri (GA/LOGOS)  
H. Werner<sup>†</sup> (FZJ)  
J. Zakova (KTH)

#### *Description*

Deliverable D121 describes the reference HTR systems, which are the starting points of the HTR core physics investigations in WP1. These reference systems are contemporary representatives of the two main HTR designs:

- Pebble-bed HTR with continuous re-load of fuel elements. This type is represented by the PBMR-400 design [1-3], although loaded with U-free fuel.
- Prismatic block HTR. This type is represented by the GT-MHR design [4,5].

As a starting point for the coated particle fuel geometry and composition, a number of *reference fuels* were defined and documented. In the course of the project the insights gained in Work Package 2 were taken into account in improved fuel designs [6-8].

Finally, D121 also presents a list of parameters to be calculated for each of the combinations of reactor system and fuel. The selection of parameters is based upon a similar list from the Euratom FP5 project HTR-N/N1 [9]. This was intended to facilitate in inter-comparison of several reactor/fuel combinations studied in this Work Package.

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## 1.2.2 HTR core physics analysis tools & opportunities for additional qualification (D111, D112)

### 1.2.2.1 HTR core physics analysis tools (D111)

#### *Authors*

J. Zakova (KTH)

#### *Description*

The list of (HTR) core physics analysis codes in use within the PUMA project contains both deterministic (WIMS, ZIRKUS, APOLLO, PANTHERMIX) and stochastic (MCNP, MCB, MONTEBURNS) computational codes. Some of the codes are focused on precise neutronics and burn-up calculations (MCB, MONTEBURNS), whereas the others allow for coupling of the neutronics and thermal hydraulics (PANTHERMIX, ZIRKUS) and therefore enable a complex investigation of the HTR systems. Some of the codes use simplifications when treating the fission products and the minor actinides (WIMS, ZIRKUS, APOLLO), whereas for example MCNP, MCB and MONTEBURNS treat explicitly all the elements. Most of the codes were benchmarked for the HTR system; some of them have experimentally validated for uranium fuels. *None of the codes has been specifically validated experimentally for HTR Pu and/or MA fuels.*

#### *Conclusions*

- The set of codes used in the investigations allows for a complex and detailed investigation of HTR, both for the prismatic block and the pebble bed design. For the pebble bed design, another approach to modelling of a real system can be employing a random distribution of the pebbles, which is available in MCNP5 [1, 2] or MONK [3].
- Although none of the codes has been specifically validated experimentally for HTR Pu and/or MA fuels, most of the codes, which do not lack the treatment of higher minor actinides, have potential to be successfully used for HTR Pu/MA investigations. It must be taken into account that simplifications of the treatment of the fission products, which result from higher minor actinides fission may lead to imprecision in calculated decay heat values. Possibilities to perform an experimental validation have been further investigated within the PUMA project (see Deliverable D112).

#### *Recommendations*

- It is recommended that appropriate experimental data be generated, on which further qualification (validation) can be based of core physics analysis codes for Pu/Ma-loaded HTRs.

- For completeness of the HTR system study we would like to propose a research on <sup>14</sup>C-production in coated particles, which can e.g. be done by the MCB-code.

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#### 1.2.2.2 Opportunities for additional qualification (D112)

##### Authors

B.Y. Petrov (NRG)

J. Oppe (NRG)

J.C. Kuijper (NRG)

##### Description

Opportunities have been investigated to obtain experimental data on which additional code qualification, aimed at the analysis of Pu/MA-loaded HTRs, can be based. A literature study has been carried out [1,2] and contacts with other Euratom FP6 projects as well as with external organisations have been established through the specific designated contact persons (ref. [3], update in D112).

Presently, the only possibility identified is the upgraded “Hot ASTRA” facility of the FSI RRC “Kurchatov Institute” in Moscow, Russia. In the project description [4], it is mentioned that experimental investigations may be defined at the modernized ASTRA facility with heating of plutonium fuelled critical assemblies up to high temperatures, provided that plutonium-based compacts/pebbles are fabricated in sufficient quantities. However, the upgrade project has not been completed yet and no experimental data has been made available within the duration of the PUMA project. *The liaison with the “Hot ASTRA” project is expected to be continued within the Euratom FP7.*

The **Low Flux Reactor** (LFR) in Petten, The Netherlands, is a 30 kW “Argonaut”-type nuclear research reactor operated by the Nuclear Research and Consultancy Group NRG. It has the unique feature that relatively large objects can be accommodated for irradiation: up to 1.10 m wide and 1.10 m high.

Some preliminary investigations have been performed to assess the potential of LFR-type reactors in the investigation of reactivity effects of advanced HTR fuel, i.e. containing Pu and/or MA. A detailed model of the **Low Flux Reactor** for use in the MCNP code [5,6] was developed at NRG, and in order to assess the reactivity effect of the introduction of a certain amount of extra nuclear fuel in the core of this reactor, a calculation test has been performed, in which a small UO<sub>2</sub>-sphere is introduced on top of the core.

##### Conclusions

- Except for the ISTC Project #685.2 “Hot ASTRA”, no opportunities have been identified which may yield experimental data on which the further qualification (validation) of core physics analysis tools can be based for the application to Pu/MA-loaded HTR systems. Even for this project the possibility to introduce Pu-based HTR fuel only exists if and when such fuel will be fabricated in sufficient quantities.
- The preliminary investigations on the Petten **L**ow **F**lux **R**eactor demonstrate its potential to accommodate experiments on reactivity (and possibly spectral as well) effects of different types of fuel (including new types of HTR fuel containing plutonium and minor actinides), which can be used for validation of calculation methods and computer codes.

### *Recommendations*

- Further investigations are recommended into the possibilities for using low/zero-power critical facilities, such as the Petten **L**ow **F**lux **R**eactor, to study the reactor physics properties of HTR Pu/MA-based fuel, as well as to yield experimental data on which the further qualification (validation) of core physics analysis tools can be based.
- It is also recommended to continue the liaison with ISTC project #685.2 “Hot ASTRA” anyway, as the experimental data expected from this critical experiment is already very useful for the further validation of core physics analysis tools for HTRs with UOX-fuel at elevated temperatures. Experimental data on Pu-loaded critical HTR configurations may become available from the upgraded ASTRA facility as well, if and when such fuel will be fabricated in sufficient quantities.

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## **1.2.3 Core physics analysis of Pu/MA HTR systems (D122 - D127)**

### **1.2.3.1 Pu and MA deep burn in pebble-bed HTR (D122)**

#### *Authors*

B.Y. Petrov (NRG)

W. Bernnat (University of Stuttgart, USTUTT)



J.B.M de Haas (NRG)

J.C. Kuijper (NRG)

A. Meier (University of Stuttgart, USTUTT)

J. Oppe (NRG)

C. Pohl (Forschungszentrum Jülich GmbH, FZJ)

### *Description*

Deliverable D122 contains conclusions and recommendations resulting from analysis of the pebble-bed high temperature gas-cooled PBMR-PUMA reactor, as described in PUMA Deliverable D121 (see section 1.3), which has been carried out by three participating organisations. Detailed results are shown in the respective Appendices of D122:

- Appendix A: “Analysis of Plutonium and Minor Actinide Utilisation in a Pebble-Bed HTR” (D122a; Nuclear Research and Consultancy Group NRG, Petten, The Netherlands);
- Appendix B: “PUMA, Work Package 1, Deliverable D122b, Part 2, calculations for pure PuO<sub>2</sub> fuel and fuel composition (PuO<sub>2</sub> + MA) in the PBMR-PUMA pebble bed high temperature reactor” (D122b; Forschungszentrum Jülich GmbH (FZJ), Germany);
- Appendix C: “Report on assessment of “deep burn” Pu/MA utilisation in pebble bed [V]HTR” (D122c; Institut für Kernenergetik und Energiesysteme IKE, Universität Stuttgart (USTUTT), Germany).

The PBMR-PUMA reactor is loaded with plutonium and minor actinide fuel and operates in a continuous reload (multi-pass) mode at nominal power of 400 MW.

Two fuel compositions have been studied by all of the partners:

- Pu – first reference fuel composition, with the isotope vector of first-generation plutonium;
- PuMA – second reference fuel composition, a mixture of plutonium and minor actinides.

Three other fuel compositions have been considered by NRG:

- Pu<sub>2</sub>xMA – fuel composition with double minor actinide content compared to PuMA, i.e. the weight fraction of Np-237, Am-241, Am-242m, and Am-243 is doubled, while the weight fractions of the plutonium isotopes are decreased proportionally;
- IMX – inert-matrix fuel with isotope content identical to the one of PuMA;
- WP – “wallpaper” fuel with the same isotope content as PuMA. In the “wallpaper” fuel, the fuel kernels are condensed to the outer diameter of the fuel zone, leaving the central part of the pebble free of fuel.

The IMX fuel features a fuel kernel diameter of 500 µm, while all of the other types of fuel have fuel kernel diameters of 200 µm. The total heavy metal loading per fuel pebble is 2 g.

Calculations have been carried out to determine the equilibrium state of the core in each case, the power density distribution and core inventory of actinides and selected fission products at equilibrium, the amount and composition of fresh and discharged fuel, temperatures at nominal conditions, temperature reactivity coefficients, control rod worth, and reactivity effects of water ingress (the latter for the first and second reference fuel compositions only).

The PANTHERMIX code system has been applied at NRG for full-core neutronics and thermal-hydraulics calculations, with a nuclear database generated by the WIMS code. The

FISPACT code has been used to calculate the nuclide inventory after a specified decay time following discharge of fuel.

The V.S.O.P.(99/05) code system has been used at Forschungszentrum Jülich.

The calculations at Universität Stuttgart were performed with the code system ZIRKUS. The thermal-hydraulic code THERMIX/KONVEK has been used for the calculation of fuel and graphite temperatures.

### Conclusions

- The reactor fuelled by Pu features higher maximum values and larger axial gradient of neutron flux (fast and thermal) and power density in the core than those fuelled by plutonium and minor actinides. In radial direction, peaks are observed at the boundaries of the inner and outer reflectors, where stronger moderation and less absorption take place compared to the core region. The pebbles in these parts of the reactor have the maximal fission power. In the Pu case, the maximum power per pebble is at least by a factor of 1.2 higher than the one in the PuMA case. The power per pebble is crucial for the temperature in the kernel. The temperature inside the kernel is much higher than the requested 1200 °C. In the Pu case, the temperature in the kernels reaches 1690 °C and in the PuMA case it is up to 1440 °C.
- The calculated attained average discharge burn-up of the different fuel types studied varies significantly between the three partners. For Pu fuel, it ranges from 747.7 MWd/kg (NRG) through 690 MWd/kg (USTUTT) to 674 MWd/kg (FZJ). The total amount of plutonium isotopes in the discharged pebbles in this case is 15.86% of their initial mass, according to the results obtained at NRG, while the total amount of actinides is reduced to 24.64%. The fraction of fissile isotopes decreases from 66.97% in the fresh fuel to 7.91% in the discharged fuel. In the calculations performed by USTUTT, the amount of fissionable material decreases to 4.54% at 690 MWd/kg. The total mass of plutonium is reduced to 21% and the mass of actinides decreases to less than 29%. Nearly all Pu-238 and Pu-239 are transmuted, only Pu-242 increases by a factor of 1.8. The total plutonium content of the discharged fuel is reduced by 77% compared to the fresh one while nearly all Pu-239 and about 57% of Pu-241 are incinerated, show the results of FZJ.
- Large differences in the average discharge burn-up calculated by the three organisations are also observed in the case of PuMA fuel. It varies from 661.3 MWd/kg (NRG) through 575 MWd/kg (USTUTT) to 431 MWd/kg (FZJ). The total amount of plutonium in the discharged pebbles in this case is 26.09% of their mass in fresh pebbles and the total amount of actinides in the discharged fuel is 33.26% of the initial mass, according to the results obtained at NRG. The fissile fraction decreases from 58.21% in the fresh fuel to 15.07% (NRG) or 10% (USTUTT) in the discharged fuel. USTUTT reports that the total mass of Pu in this case decreases from 89% to 30% and the mass of all actinides decreases to 40%. The amount of Pu-238 increases a little bit due to the n-capture in Np-237 which is present in the fresh fuel. Pu-240 stays at nearly the same value as in the fresh fuel and Pu-242 increases as in the Pu case. The amount of Pu-239 is reduced by 87%, Pu-241 shows a build-up to 144% of the initial isotope mass, while the total plutonium content of the discharged fuel is reduced by 48%, according to FZJ.
- In the case of Pu2xMA fuel, the achievable average discharge burn-up is 378.1 MWd/kg and then the plutonium content in the discharged pebbles is 59.17% of their initial mass. The total amount of actinides is reduced to 61.82%. The fissile fraction decreases from 51.01% in the fresh fuel to 36.42% in the discharged fuel.

- The fuel of IMX type is able to attain average discharge burn-up of 692.6 MWd/kg. The total amount of plutonium in the discharged pebbles is reduced to 22.06% of their initial mass and the total amount of actinides is reduced to 30.11%. The fissile fraction decreases to a level of 10.10% in the discharged fuel.
- The total amount of plutonium in the discharged pebbles of WP type (with average discharge burn-up of 656.1 MWd/kg) is reduced to 26.74% of their initial mass. The total amount of actinides is reduced to 33.79% and the fissile fraction decreases to 15.88% in the discharged fuel.
- These results demonstrate the excellent plutonium and minor actinide burning capabilities of the high temperature reactor. The largest degree of incineration is attained in the case of an HTR fuelled by pure plutonium fuel as it remains critical at very deep burn-up of the discharged pebbles. Addition of minor actinides to the fuel leads to decrease of the achievable discharge burn-up and therefore smaller fraction of actinides incinerated during reactor operation. The inert-matrix fuel design improves the transmutation performance of the reactor, while the “wallpaper” fuel does not have advantage over the standard fuel design in this respect.
- After 100 years of decay following the fuel discharge, the total amount of actinides remains almost unchanged for all of the fuel types considered. Among the plutonium isotopes, only the amount of Pu-241 is reduced significantly due to its relatively short half-life. Slight build-up of Pu-239 and Pu-240 is observed due to the decay of Am-243 and Cu-244.
- The total temperature reactivity coefficients at Cold Zero Power (isothermal reactor at 300 K, all control rods out, and zero xenon conditions) are positive in all of the cases. The total temperature reactivity coefficients at Hot Zero Power (isothermal reactor at nominal coolant inlet temperature of 773.15 K, all control rods out, and zero xenon conditions) are slightly positive or slightly negative, with the exception of the case of Pu2xMA fuel, where the strong negative temperature reactivity coefficient for fuel and moderator assures a negative total temperature reactivity coefficient. In the case of Hot Full Power state (temperature distribution at nominal conditions, control rods at critical position, and equilibrium xenon conditions), the large negative temperature reactivity coefficient for fuel and moderator compensates for the slightly positive reflector coefficient and this yields a negative total temperature reactivity coefficient.
- The reactivity increase by withdrawal of all control rods would be around 2.9% in both Pu and PuMA cases, and such a value of the control rod worth is considered too high by USTUTT. This represents an increase of more than 7\$ which is too much in respect to reactivity accidents (e.g. by withdrawing of all control rods starting from an equilibrium position). The xenon reactivity effect is much smaller for plutonium-based fuel compared to the one in case of uranium-based fuel.
- Finally it should be noted that the PBMR-PUMA reactor and fuel composition data which have been transferred from PUMA WP1 to WP3 (Milestone M122) for use in the DANESS code (see PUMA Support Document S128) are identical to the data presented in Appendix A of D122 in case of fresh fuel, and differ by 3% to 4% (measured as average relative error for all of the actinides) in the cases of the highest burn-up values considered. Therefore, the conclusions drawn in PUMA WP3, based on these data, remain valid.

### *Recommendations*

Based on the conclusions drawn from the analysis of the pebble-bed high temperature gas-cooled PBMR-PUMA reactor, the following recommendations can be given:

- *Reduction of the plutonium load per pebble.* The under-moderated core causes a strong increase of reactivity at water ingress. Calculations with a reduced plutonium load performed by FZJ suggest that for an amount of 0.75 g plutonium per pebble instead of 2 g the increase of the reactivity at water ingress is limited to a maximum of about 1 Nile at 120 bar water pressure.
- *Reduction of the power to approximately half of the value for the current design or change of the reactor geometry.* These are needed in order to keep the temperature in the fuel kernels within the safety limits.
- *Establishment of procedures of bringing the core into a hot state in a controlled manner or changes in reactor design.* This recommendation is derived from the results for the temperature reactivity coefficients at Cold Zero Power and Hot Zero Power states.
- *Reduction of the number of control rods.* This could solve problems associated with too high control rod worth. Alternatively the design of the control rods could be changed to reduce the worth.
- *Validation of the computer codes used by various organisations.* Considerable differences have been observed between results obtained by the three partners within this subtask of PUMA WP1, which however are not expected to invalidate the overall conclusions based on the analyses. Among the possible causes for that are e.g. different handling of epithermal Doppler broadening and modelling of resonance interference, different nuclear data libraries and values of energy release per fission used, or differences in the thermal-hydraulic models. Further investigation is necessary in this regard. Code validation through experimental data or reference calculations is needed, and such an activity might be subject of a future project (also see sections 1.2.2.1 and 1.2.2.2).

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### 1.2.3.2 Assessment of Th/Pu fuel cycle in pebble bed [V]HTR (D123)

#### *Authors*

C. Pohl (FZJ)

H.J. Rütten (FZJ)

#### *Description*

FZ-Jülich started research on the use of Plutonium/Thorium and Pu/U/Th-based fuel in Pebble-bed-HTRs early in the last decade of the past century [1,2]. This work was continued within the frame of a Coordinated Research Program (CRP) of the IAEA from 1995 through 2001 [3,4].

The incineration of Plutonium of first and second generation was investigated in the course of the EC-funded Projects HTR-N and HTR-N1. A list of related publications is given in refs. [1-5].

Recent FZJ activities within the PUMA project concern a fuel composition of plutonium with thorium, which is intended to satisfy the safety regulations for a high temperature pebble bed reactor. To combine this request with a high burn-up of the total plutonium content of the fuel

and in particular the amount of the fissionable isotopes the fuel composition should be optimised.

The combination of plutonium with an additional amount of thorium has in general many positive effects as the reduction of peak fuel temperature, power per pebble, power density and because of the Doppler broadening at higher temperatures a negative temperature reactivity effect. In addition the breeding effect generates an additional amount of the fissionable isotope U-233. As a negative effect should be mentioned that the additional mass of thorium increase the total heavy metal load per pebble which reduce the moderation ratio to an under moderated stage and therefore the criticality of the core.

The numerical calculations of the critical equilibrium and other core conditions with V.S.O.P. (99/05) take into account the resonance broadening and self shielding of thorium and plutonium Pu-242.

### *Conclusions*

- For a kernel size 0.05 cm in diameter a content of 2 g first generation LWR plutonium and 7 g of thorium nearly satisfy the safety request for a negative total temperature coefficient for the tree considered core conditions at the expense of a relative low total burn-up of about 51 MWd/kgHM and a plutonium specific burn-up of about 230 MWd/kgPu. This individual burn-up corresponds to an incineration of about 29 % of the initial plutonium. But for this composition a massive increase of the reactivity at water ingress occurs which reflects the very poor moderation ratio for such a high heavy metal load of the pebble.
- A lower plutonium load of about 1 g per pebble can handle a significant higher amount of thorium with a coincidentally high individual burn-up of about 650 MWd/kgPu which corresponds to an incineration of about 70 % of the initial plutonium. Also the increase of the reactivity at water ingress is much lower and has in case of 1g thorium per pebble almost no influence. On the other hand this amount of thorium is not enough to compensate the positive moderator and reflector temperature reactivity coefficient, in particular for a cold isothermal core.
- The main problem of the temperature reactivity coefficient for a fuel composition with low amount of thorium arises from the isotope Pu-241. With a harder thermal spectrum this results first in an increase of the moderator coefficient up to a maximum. In combination with the also positive reflector coefficient a high total positive coefficient will generated. For a still higher temperature the moderator coefficient decreases down to a negative value just as the total temperature coefficient.
- A modification of the kernel size from 0.05 cm in diameter down to 0.02 cm results in a slightly higher burn-up and a significant better temperature reactivity coefficient but is still not enough to satisfy the safety requirements.

### *Recommendations*

- A combination of plutonium with a limited thorium mass in a small kernel can result in an optimised fuel for deep burn-up of plutonium. The still positive temperature reactivity coefficient of a cold zero power core should be changed, and also the heterogeneous flux and power density distribution with the hot spots in vicinity of the inner and outer reflector.
- Additional calculations for fuel mixtures of plutonium, thorium and minor actinides are useful and necessary as well as transient calculations on accidental water ingress.

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### 1.2.3.3 Pu/MA (“deep burn”) utilisation in prismatic [V]HTR (D124)

#### Authors

J. Cetnar (AGH)  
Yonghee Kim (KAERI)  
J.T. Murgatroyd (AMEC/NNC)  
C. Trakas (AREVA)  
F. Venneri (GA/LOGOS)  
J. Zakova (KTH)

#### Description

This section summarises the work done under the scope of Deliverable D124 for the PUMA project, which is part of the European Commission’s 6th framework program. It includes contributions from AGH, KTH, NNC, LOGOS and AREVA NP, which are presented in the respective Appendices of D124:

- Appendix A: Assessment of Pu and MA utilisation in deep burn Prismatic HTR by Monte Carlo Method - MCB (AGH)
- Appendix B: MCNP/MCB analysis of GT-MHR (KTH)
- Appendix C: Modelling of the PuMA Prismatic Core with a Coupled Neutronics and Thermal Hydraulics Code Scheme (AMEC/NNC)
- Appendix D: Design Optimization of the 600MW<sub>th</sub> Deep-Burn MHR Core (GA/LOGOS)
- Appendix E: Optimization of the Deep-Burner-Modular Helium Reactor (DB-MHR) concept for actinide incineration (AREVA NP)

The main mission of PUMA reactor development is to design a safe and effective plutonium and minor actinide burner reactor. On the road to that goal several design decisions need to be taken basing on the reliable analysis of reactor core physics in deep burn mode. The effect of deep burn on core physics involves several dependences, which complicate analysis, and in case of model simplification can lead to biased results. Primarily, it is bounded with power

spatial distribution and its time evolution due to burn-up, fuel shuffling and reactivity control. The identification of that dependences and accurate assessment of their meaning and influence on the core performance are the applied calculation model reliant. Yet, calculation models employ different nuclear data libraries, their different implementation or treatment in the model, which in case of deep burn design can amplify differences in the results. Therefore, core design analysis performed with different tools is very important, and can lead to recognition of existing problem in applied design tools and models. Analyses of PUMA core performance for Deliverable D124 have been carried out with application of diversified tools and models. This summary report comprises an informative description of applied design tools and models, the most important findings, discussion and recommendations.

AGH and KTH have studied the problem using Monte Carlo MCNP[1] and MCB[2] codes in the models with complete description of double heterogeneity. The AGH study has involved thermal-hydraulic coupling with POKE[3] and a novel “bridge scheme of burn-up step” to reduce the flux oscillations and convergence problems. Continuous energy cross sections have been applied there that are based on JEFF3.1 library. AGH has modelled the CR operation to investigate its influence of core performance.

NNC has used the deterministic approach where the calculation route consists of a WIMS[4] neutronics model, a bespoke thermal hydraulics code and a Python script to couple the two. The overall model consists of a whole-core diffusion model and several cell models, which are used to calculate material average group constants for the whole-core model. The cell model calculations are performed in 172 energy groups using the standard WIMS library (based on JEF2.2). The model is a simplified one with no representation of control rods and cooling channels in the axial reflectors.

The GALOGOS has used a Monte Carlo and deterministic hybrid method – the HELIOS[5]/MASTER-GCR[6] code system. It includes a thermal-hydraulic-coupled neutron analysis. In this method the double-heterogeneous fuel compact is transformed into a conventional single-heterogeneous problem and then the conventional two-step core analysis is done. The cross section libraries have been generated from the ENDF-B/VII data.

AREVA has applied MCNP-MONTEBURNS-ORIGEN chain, with either JEF2.2 or BVI libraries in simple models that has used 2D and 3D geometry models but with simplifications in the core description.

The results of the core performance concerning the plutonium and actinides destruction are very similar in all applied methods. The major differences concern achievable burn-up, power distributions and reactivity swing, which are attributed to the applied methodology and models, in regard to power and burn-up distributions and the influential CR modelling.

The total actinide consumption rate in the core was found to be 41.8 kg/TWh(th), which is dominated by the plutonium consumption rate of 41.0 kg/TWh(th). This in turn is dominated by the <sup>239</sup>Pu consumption rate of 32.3 kg/TWh(th). Most of the remaining contribution is from <sup>240</sup>Pu with a consumption rate of 13.0 kg/TWh(th). Since <sup>240</sup>Pu has a low fission cross section compared to the odd isotopes of plutonium but a high capture cross section, the main mechanism for its consumption is probably capture to <sup>241</sup>Pu. This would also explain why the net consumption rate of <sup>241</sup>Pu is quite low, at 1.9 kg/TWh(th), even though it has a relatively high fission cross section at thermal energies. The core is a net producer of <sup>238</sup>Pu and <sup>242</sup>Pu with production rates of 2.7 kg/TWh(th) and 3.5 kg/TWh(th) respectively. The consumption rate of <sup>237</sup>Np is also reasonable, at 3.0 kg/TWh(th), considering that its start of life inventory is so much lower than that of <sup>239</sup>Pu. Again, this is probably due to capture rather than fission, resulting in the comparatively large net production rate of <sup>238</sup>Pu. The net consumption rate of americium is rather small at <0.2 kg/TWh(th), although this is partly due to the small start of



life inventory. It consists of a consumption rate  $\sim 1.4$  kg/TWh(th) for  $^{241}\text{Am}$  and a production rate  $\sim 1.2$  kg/TWh(th) for  $^{243}\text{Am}$ . Like  $^{237}\text{Np}$ ,  $^{241}\text{Am}$  has a rather low fission cross section at thermal energies, so the main means of consuming it is via capture to  $^{242}\text{Am}$ , which decays very rapidly from its ground state to  $^{242}\text{Cm}$ . This mechanism for removing  $^{241}\text{Am}$  has to compete with the production mechanism of decay from  $^{241}\text{Pu}$ . There is a net production rate of 2.3 kg/TWh(th) for curium, due to captures in americium. This is inevitable in a reactor that is designed primarily for burning plutonium and americium. There is also a small uranium production rate of 0.1 kg/TWh(th).

The results of actinide consumption quoted above have been obtained by NNC for the 2<sup>nd</sup> reference fuel. Similar results obtained in other evaluations can differ by a small deviation up to 5% (total actinide consumption rate in AGH evaluations of a similar core equals 44.42 kg/TWh<sub>th</sub>), which probably can be attributed to different power normalization or the core modelling features.

### Conclusions

- Achievable burn-up of 65% FIMA for 1<sup>st</sup> reference fuel and 55% for 2<sup>nd</sup> reference fuel has been found in MCB Monte Carlo model of AGH investigation. These values have been generally confirmed in GA/LOGOS and KTH assessments, whereas the deterministic model of NNC has given a higher number (concerning 2<sup>nd</sup> reference fuel). The difference of about 20% is probably caused by applied model simplifications like the lack of CR modelling or different treatment of cross sections.
- Burn-up reactivity swing varies significantly depending on the methodology or applied burnable poisons. It ranges from 1800pcm – 5700pcm (not including the swing related to Xe135). In every case, the compensation with CR is manageable. The reactivity swing has a limited influence on burn-up, since the cores with suppressed reactivity swing have lower criticality level at BOL.
- Operation of CR significantly influences axial distribution of power and burn-up and reverses the effect of the temperature impact on the power profile.
- The modelling of CR operation is necessary for a proper evaluation of axial power profile and power peaks. This assessment done in AGH investigation has been not carried out in other investigations, therefore different power distributions are obtained.
- Power peaking can be controlled effectively with axial-only shuffling scheme, with additional power suppression in fresh fuel block by burnable poison.
- Power peaks during fuel cycle occur when operational CR insertion level is raised above the block layer with fresh fuel.
- Statistical oscillations in Monte Carlo solution of transport equations in PUMA fuel cycle have been reduced by “bridge scheme” of burn-up step.
- Temperature reactivity coefficients are negative in all conditions in AGH study with  $\text{Eu}_2\text{O}_3$  poison, but in CZP the graphite coefficient is positive, with no safety threat however.
- Detailed investigation of reactivity in GA/LOGOS investigation shows that both  $\text{B}_4\text{C}$  and  $\text{Er}_2\text{O}_3$  are promising burnable poison material for the DB-MHR core and  $\text{B}_4\text{C}$  provides a little higher fuel discharge burn-up. However, the isothermal temperature coefficient of the core can be positive with the  $\text{B}_4\text{C}$ , while it can be strictly negative with  $\text{Er}_2\text{O}_3$  over the whole range of core temperature.

- Plutonium and MA can be effectively utilised in deep burn prismatic HTR, while conserving the safety constraints.

#### *Recommendations*

- Additional development and improvement of design tools and models is needed in order to design appropriately a prismatic HTR dedicated for deep burning of plutonium and minor actinides.
- A benchmark activity is required, devoted to the evaluation of prismatic deep burn HTR core performance with different approaches.
- In core performance optimization the CR operation must not be neglected since it impacts the power and burn-up distribution.
- Power profile peaking in the reflector vicinities need to be assessed in a finer radial mesh than the thickness of one block, in order to avoid power peaking underestimation.
- Burnable poison needs to be used for power peaking reduction. A better optimization is needed with the consideration of CR operation.
- In order to accomplish an effective optimization of the core design there is required well-defined set of constraints that needs to be formulated yet. This should also include analysis of decay heat and possible cooling accident scenarios.

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#### 1.2.3.4 Assessment of Plutonium-Thorium fuel cycle in Prismatic [V]HTRs (D125)

##### *Authors*

E. Girardi (EDF)  
J. Cetnar (AGH)  
M. Kopec (AGH)  
D. Lecarpentier (EDF)

##### *Description*

Compared to the other Gen IV reactor concepts, HTRs do not authorize high breeding ratios when relying on Uranium or Plutonium fuel. Consequently, the requirement of sustainability with respect to the use of fissile resources may be questionable, especially if the demand for high-temperature applications grows significantly in the next century.

This document investigates the feasibility of a prismatic [V]HTR Th/Pu fuel cycle with a high conversion ratio (around 0.8). This concept is designed for an efficient burning of the Pu in conjunction with high Th-conversion and U233-burning rate.

Various computational models were used in this work:

- Some simplified models (e.g. 1D and 2D models), used in the preliminary analysis context;
- A “reference” deterministic model, based on the lattice code APOLLO2, the core code Cronos2 within the HTR reactor analysis package NEPHTIS;
- A 3D Monte Carlo model, based on the depleting Monte Carlo code MCB (neutronics) and the POKE code (thermal-hydraulics).

In order to optimize the Pu burning and fuel conversion rate in the Th/Pu fuel cycle, several design parameters were considered in this work:

- Fuel composition Pu/Th, Pu/Th/U3, U3/Th
- Heavy Metal total mass in the core
- Fuel packing fraction and particles size
- Core layout: annular (reference GT-MHR, with 3 fuel rings or DB-MHR, with 5 fuel rings) or cylindrical (8 fuel rings, without inner reflector)
- Uniform and non-uniform Thorium distribution and the associated shuffling scheme

### Conclusions

- In D125 we show that use of thorium is promising regarding the breeding ratio. Moreover the symbiotic Pu/Th fuel cycle is a good means to control the Pu inventory and optimize the energetic use of plutonium relying on the thorium conversion into fissile uranium.
- The fuel performance was optimized by maximizing the fuel conversion rate with a constraint over the cycle length. It is shown that an acceptable trade-off among these two key parameters can be met when considering a very high Heavy Metal mass and a large Thorium fraction in the fuel composition.
- Using to the modified DB-MHR core layout (e.g. DB-MHR core, where the inner reflector has been replaced by standard fuel columns) and about 24tHM for a fuel composition of 97%Th – 2.4%U<sup>233</sup> – 0.6%Pu, an acceptable 265 full power days operating cycle was reached. Such a design results in a U<sup>233</sup> self-breeder concept where the Pu consumption is limited to 20 kg Pu per TWhe.
- Also, a preliminary safety analysis, focusing on feedback coefficients was performed. It showed good reactor features (Doppler –4 pcm/°C, moderator ~0 pcm/°C, reflector +0.25 pcm/°C). Moreover, results obtained on core power distribution and Control Rod worth comply with the operating constraints, even if some further optimization are required.
- Additionally, very promising results were obtained for non-uniform Th distribution – especially when Th was located in the 2 innermost fuel rings wrapping the inner reflector. Considering a more complex optimum search (including cylindrical configurations with

heterogeneous Th/Pu distribution), one can expect to get better conversion rate and larger cycle length.

- Moreover, an interesting feature of the heterogeneous fuel management is related to the small amount of  $U^{232}$  produced, while the  $U^{233}$  is extracted from blocks without Pu fuel. In this context, reprocessing of Th kernels would be easier as the Pu and Th are physically separated. Compared to the uniform Th distribution cases, this may be preferable despite of lower production.

### *Recommendations*

- Although viable from the neutronics point of view, the proposed Pu/Th modified DB-MHR core needs a more extensive validation, due to the core layout modifications (removal of inner reflector, modified thermal-hydraulics conditions, and lower power density) with respect to the standard DB-MHR reactor and needs to be reconsidered from the economic point of view. Indeed, large cores need larger amount of heavy metals and fissile materials.
- Also, further optimization/design work is required for a better assessment of Pu/Th fuel cycles in [V]HTRs :
  - Optimization of the power shape, by means of burnable poison
  - Optimization of the CR weight and position, in order to deal with larger fuel cores (e.g. DB-MHR core, where the inner reflector has been replaced by standard fuel columns)
  - Explore the feasibility of the non-uniform Th distribution in a Large Core (e.g. DB-MHR core, where the inner reflector is replaced by Th fuel columns)
  - Investigate the possibility of an even more non-uniform Th distribution: mixing Th and Pu compacts within the same Fuel Assembly.
- In addition, the use of thorium requires significant changes of the fuel cycle and the associated fuel plants. It also requires the ability to reprocess, in an industrial context, the HTR fuel micro-particles in order to extract uranium and thorium and using them in a multi-recycling context. Due to the presence of  $U^{232}$  and its daughters in the fuel, reprocessing, production and the fuel transportation will be more complicated and always associated to highly emitting materials. All these points have to be addressed.

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#### 1.2.3.5 Reactivity transients of Pu/MA fuelled [V]HTR (D126)

##### *Authors*

A. Meier (USTUTT)

W. Bernnat (USTUTT)

K. Hossain (USTUTT)

##### *Description*

Analyses on a PBMR – reactor – design were done with the assumptions made in D121 [6]. For two different kinds of fuels (the first one with Pu, the second one with Pu, Np and Am) an

equilibrium core was calculated. The results were evaluated with respect to temperature coefficient, water ingress and slow and fast reactivity transients (e.g. control rod ejection).

The stationary design calculations were done with program system ZIRKUS. Eleven energy groups are used (6 fast (till 2.38 eV) and 5 thermal) for the solution of the neutron diffusion equation. The thermal-hydraulic program for stationary states was THERMIX which was extended in a way that the temperatures in the kernel for every pass of the pebbles in the reactor can be calculated. The transient calculation was done with an in-house three dimensional thermal-hydraulic program TH-3D coupled with point-kinetics.

All results have been compared with the PBMR uranium benchmark as well [7].

### *Conclusions & recommendations*

- For the first reference fuel (pure Pu) a critical core is reached with 690 MWd/kg HM. For the second reference fuel only a discharge burn-up of 575 MWd/kg HM can be reached. This is due to the higher amount of nuclides with a higher absorption already in the fresh pebbles.
- The power is strongly peaked on the inner and outer reflector boundary. This can be explained with the high under-moderation in the reactor and the strong moderation part of the reflector. In these parts of the reactor the pebbles will have the maximal power. In the Pu-case the power per pebble is a factor of 1.2 higher as in the PuMA-case. The power per pebble is crucial for the temperature in the kernel. The temperature inside the kernel is much higher as the requested 1200 °C. In case of Pu the temperature in the kernel will reach 1689 °C and in the PuMA-case 1438 °C.
- The moderator temperature coefficient in the Pu-case is positive below 450 °C. Also the sum of fuel and moderator coefficient is slightly above zero. The same, even if a little better, is the case for the second reference fuel. In the uranium case, moderator and fuel coefficient are always below zero and their sum is always below  $-5 \times 10^{-5} \text{ K}^{-1}$ .

Due to the high temperature, the heterogeneous power distribution and the positive temperature coefficient, some design modifications were tested.

- Mass reduction (1 g HM per pebble) leads to even higher temperatures and to stronger positive moderator temperature coefficients.
- Power reductions (200 MW instead of 400 MW) can be an improvement for the temperatures, but the temperature coefficients are not affected.
- Changing the geometry to a fully cylindrical core geometry with 1.5 m radius results in a much better power distribution and also in much lower fuel temperatures. But the moderator coefficient is still positive around 300 °C, but becomes stronger negative for high temperatures (> 1000 °C).
- Using a two-zone-geometry with 1.85 m radius, where in the outer part the fresh pebbles and in the inner part the already burned-up pebbles are loaded, one observes also a good result, even if the power distribution is very heterogeneous. A change in loading (fresh in the inner part and burned-up in the outer part) will show better results and can be a real alternative. But of course one has to look more in detail.
- The water ingress is only calculated in a stationary way. For all mentioned designs, the water ingress leads to a positive reactivity. The best case would be the 1.0 g HM in the pebble, of course because of the lower amount of heavy metal in the core. But due to strong under-moderation, water ingress improves the moderation (up to optimal moderation) and the reactivity will increase.

- Calculating the depressurized loss of coolant (DLOCA) accident we observed for all analysed cases that the maximum temperatures are lower than 1600 °C. Because it is a slow transient with very low power density, the temperature difference between the fuel and the graphite is unimportant.

For the reactivity transients, control rod ejections with different velocities were simulated. For the slower transients, it was assumed that the control rods move out with only 1 cm/s, which is the nominal velocity. But also fast transients with 1 m/s were regarded.

- Compared with the uranium case, the power increase for Pu and PuMA is not as high as in the uranium case, but this is due to the higher temperatures, the Pu and PuMA kernel get. Due to the negative temperature coefficient, the feedback to the reactor is much faster, and therefore the power will not increase that much. Also the decrease after reaching the maximal temperature in the kernel is much faster in the Pu and PuMA case, due to the strong moderator coefficient.
- For the fast reactivity transients all 3 cases (Pu, PuMA and U) show maximum particle temperatures are above 1600°C. This must be avoided e.g. by an appropriate excess reactivity bound in control rods at nominal power. On the other hand it should also be noted that, in reality, there is no conceivable mechanism by which control rods could be "ejected". However, these transient calculations are very useful anyway to assess the consequences of a fast reactivity insertion.
- In summary: except for water ingress no significant difference in transient behaviour was found compared with UO<sub>2</sub> cores, also not for DLOCA and PLOCA accidents. But for low temperatures the moderator coefficient is slightly positive. Using a pebble bed reactor we propose to avoid the inner reflector column.

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### 1.2.3.6 Assessment of LWR-HTR-GCFR integrated fuel cycles (D127)

#### *Authors*

E. Bomboni (CIRTEN)

N. Cerullo (CIRTEN)

G. Lomonaco (CIRTEN)

#### *Description*

CIRTEN analyzed and assessed the potentialities of an integrated LWR-HTR-GCFR fuel cycle within the PUMA Specific Targeted Research Project (STREP) sponsored by the European Commission via its 6<sup>th</sup> Framework Programme (FP6).

The integrated LWR-HTR-GCFR basically aims at closing the current nuclear fuel cycle: in principle, thanks to the unique characteristics of Helium coolant reactors (thermal and fast), LWR Spent Nuclear Fuel (SNF) along with Depleted Uranium become valuable material to produce a lot of energy. Additionally, burning Heavy Metals (HM) of LWR SNF means not only a drastic reduction in the natural U demand, but also a remarkable decrease in the mass of the long-term radiotoxic component of nuclear waste to be geologically stored.

D127 is subdivided into three parts.

The first part outlines briefly the main characteristics of both the High Temperature gas-cooled Reactor (HTR) and the Gas-Cooled Fast Reactor concepts. Then the advantages of linking LWR, HTR and GCFR in a symbiotic way are underlined, stressing how much this cycle is suitable for reaching the Gen IV goal of sustainability and proliferation resistance.

The second part shows the cores we considered to assess this integrated cycle, i.e. the Pebble-Bed Modular Reactor 400 MW<sub>th</sub> as HTR concept, and the GCFR “E” 2400 MW<sub>th</sub>. A brief overview of the main technological constraints concerning Pu- and MA-based advanced fuels is given. This part is able to explain and justify the choices made in the framework of the considered cycle. Thereafter, calculations performed and results obtained are described.

Finally, the third part summarizes the most significant results. Some conclusions are drawn and future work needed is suggested. This is listed below.

#### *Conclusions & recommendations*

The analysis performed in the framework of the EU project PUMA substantially confirms previous findings, but adding some remarkable developments. Once again, as found out in previous researches, the integrated LWR-HTR-GCFR shows very good capabilities to achieve a sustainable nuclear fuel cycle. As stated before, LWR-HTR-GCFR allows us the possibility to meet the following criteria:

- Excellent exploitation of U resources intrinsically coupled with the strong reduction of the mass of the final waste (ratio waste/energy decreases by two orders of magnitude compared to OTTO).
- Reduction of the radio-toxicity of the final waste to that of FPs and MAs (i.e. reduction of LOMBT by an order of magnitude compared to OTTO).
- Strong reduction of Pu (and, if necessary, Np) stockpiles thanks to HTR loaded with fertile-free fuel, in parallel with an important change of its isotopic composition, which becomes extremely proliferation resistant (fuel that contains high masses of strong heat- and neutron-emitters).



- Very long fuel cycles thanks to the very favourable neutron economy that is typical of He-cooled reactors.

Additionally, the strategy here developed permits us some others improvements:

- Heterogeneous recycling of SNF (separating FPs, Am and Cm from U, Np and Pu) in principle removes any technological limit to the possibility of recycling HMs many times. Indeed pernicious nuclides (in small amounts anyway) like Cm isotopes are stored as a final waste or recycled in dedicated facilities.
- Multiple recycling of GCFR SNF coupled to removing Am, Cm and FPs shows the following important trends:
  - Am and Cm amounts increase even slower from cycle to cycle;
  - As a consequence, the LOMBT of GCFR SNF (Am, Cm and FPs) decreases from cycle to cycle;
  - Pu isotopic composition is good enough to sustain very long irradiation cycles (more than 9 years in principle), but it is not suitable for military uses (high content of Pu<sup>240</sup>);
  - Np is always burnt by both HTR and GCFR;
  - Pu<sup>238</sup>, which is a strong alpha- and neutron-emitter and then which could be pernicious for reprocessing, decreases during multiple recycling.
- Radio-toxicity of GCFR SNF, after less than 100 years, is essentially due to Am nuclides instead of Cm ones: that indicates designing of a dedicated assembly to burn Am, along with an opportune cooling time of SNF, as a straightforward way to close the cycle effectively.
- The excellent GCFR neutron economy and its huge DU inventory permit us the possibility to insert some dedicated assemblies to burn Am and Cm without significant consequences concerning core safety.

What's more, the analysis here performed gives us some clues regarding He-cooled reactor core modelling as well. Particularly, we found that:

- As far as PBMR modelling is concerned, burn-up calculations give results in excellent agreement with each other regardless the kind of model chosen to describe the core itself. Then, using very rough models like an infinite lattice of pebbles can be considered a good approximation, if we are only interested in the final isotopic composition. Conversely, dynamic parameters are very badly estimated by approximated models, thus we cannot rely on results supplied by this kind of calculation.
- Both PBMR and GCFR isotopic inventory vs. burn-up seems to be only slightly influenced by the axial temperature profile that can be described with Monteburns 2.0 (i.e. varying only the temperature of the non-burnable materials), so that it is not worth increasing calculation time because of a 2-zone model. Thus, we can adopt 1-zone models to simulate irradiation histories of these cores, having a higher calculation speed as a benefit.
- On the other hand, GCFR reactivity parameters are also strongly influenced by axial temperature profile, so that we cannot rely on the results found with our approximated models.
- As a future work, to close this cycle effectively, designing a dedicated assembly for Am and Cm burning is requested. Of course, a fuelling scheme for the GCFR core is

necessary as well. However, results found here seem to be very promising as far as finding this final solution is concerned.

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## 1.2.4 Additional investigations (D131, D132)

### 1.2.4.1 Prediction of Helium production in coated particles (D131)

#### *Authors*

J. Cetnar (AGH)

G. Domańska (AGH)

M. Kopeć (AGH)

S. Shihab (BN)

#### *Description*

Prior to any modelling of helium behaviour in fuel particles, the production itself of helium must be calculated. The existing experience with LWR fuel [1] is used here only for comparison purposes, since the neutronic conditions as well as fuel design of the HTR reactor with Plutonium and MA fuel significantly differ from the one of LWR. The complexity of burn-up process requires application of an integrated calculation system, which will allow taking into consideration of spatial effects of full heterogeneous reactor model with continuous energy representation of cross section and the thermo-hydraulic coupling.

We have used integrated burn-up calculation code MCB [2], in two full heterogeneous reactor models. The first one called static, is a general model that brings about helium production dependence on various combinations. It has been devoted for application in a general case of PUMA core. However, the radial distribution of burn-up in this model is characteristic to cores with fuel loading or shuffling scheme that loses memory of the burn-up spatial distribution. Such conditions occur in the pebble bed reactor because of the pebbles mixing process, which loses the local memory of burn-up radial distribution. In case of a prismatic core with axial shuffling, the axially integrated burn-up can have different distribution of power and burn-up profile. For this case, we have performed calculations in the full core burn-up model with four-batch axial-only block shuffling, according to recommendation from [3].

Helium production rates were calculated as a function of time as well as of burn-up in representative points of the core. In all cases, the highest helium production occurs in the locations closer to the core middle plane, as expected. Comparison between upper and bottom locations shows that higher production rates are observed in the upper part, which probably is an effect of higher fuel temperature there. Comparison of the production rates along radial locations one can notice that they are close in the reactor middle plane, whereas visible difference of more than 10% is noticed in the upper on bottom locations. The helium production as a function of burn-up strongly depends on the burn-up rate, not merely the

accumulated burn-up. The helium production rates in upper and bottom locations are higher than in the central locations. Yet, helium cannot accumulate to higher values there due to significantly lower burn-up achievable, as compared with central locations. This effect of higher reaction rate with lower burn-up rate leads to reduction of the spread in the accumulated helium production depending on the locations. Alpha decay of heavy metal nuclides is an overwhelming contributor to helium production in all locations. This term is responsible for differences in helium production depending on locations. Second contributing term of ternary fission is, due to its nature, proportional to the burn-up but its contribution is below 10%. The alpha emission from oxygen is very small due to deeply moderated neutron flux, which only fractionally reaches the reaction threshold of few MeV.

In order to understand what physical process is responsible for observed dependence in helium production in the PUMA core we have looked at characteristics of transmutation process that leads from the fuel to helium. This analysis shows what the reasons are of the fuel burn-up not being a proper measure of helium production. In the evolution of a fissionable nuclide, the higher flux imposes higher total reaction rate, which comprises fission and neutron capture, thus enhancing both burn-up and transmutation to an alpha emitter. Yet, in lower burn-up rates the process of alpha emitter formation, particularly of Am241 is more efficient. This way a residence time in the reactor plays an important role in flattening the resultant accumulated helium on discharge. In the static model, the radial burn-up distribution is peaking at locations close to the reflectors. Similar conditions occur in the pebble-bed reactor because of the pebbles mixing process, which loses the local memory of burn-up. The highest values of helium production on discharge occurs when fuel is kept in the reactor core for a long residence time under relatively lower burn-up rate in order to reach a target burn-up, as it is generally applicable to the pebble bed fuel.

The results of helium production on discharge of fuel that has been shuffled axially confirm the results of the previous model in that they remain in the range determined earlier for a general case. As expected, the radial distribution of burn-up on discharge is lower on the active core sides than in the radial middle locations. The helium production and the burn-up are peaking at the core middle location of the sixth block row. The core with axial shuffling is free of power peaking at the radial borders and the radial power profile is flattened. This leads to elimination of extreme values of helium production achievable in pebble bed core obtained in the relevant burn-up static model. In the current case, which is closer to a real loading scheme in a prismatic core, there is no possibility to keep fuel block for a long residence time under relatively lower burn-up rate in order to discharge fuel on a target burn-up. Thus the irradiation pattern possible in pebble bed cores, in which it the highest amount of helium accumulate, are excluded here, since the residence time of fuel blocks is the same for all blocks within the same batch load. Current results of burn-up full model remain in very good agreement with the ones obtained in the static model.

### *Conclusions*

- Helium production rate in a coated particle significantly depends on its location in the core.
- The most strongly contributing process is the decay of HM alpha emitters; ternary fission contributes only few percent while contribution from oxygen is very small.
- Higher burn-up rate lowers helium production rate in terms of burn-up.
- Extension of the residence time in the core in order to meet the burn-up target will result in higher build-up of helium.
- Higher temperature (in lower core locations) influence burn-up rate adversely but increases helium production rate versus burn-up

- The same range of helium production rates as in LWR case occurs in HTR in corresponding burn-up
- The maximum helium production occurs in pebbles that pass the core always through the middle locations with lower than average burn-up rate.

#### *Recommendations*

- "Axial only" shuffling scheme is a good solution in terms of levelling the helium production.
- Accumulation of helium in prismatic core can be reduced by increasing the burn-up rate, resulted for example from lowering fuel packing fraction.
- Burn-up control of the pebbles might be not sufficient in sense of controlling the helium content. The residence time should be also controlled.

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#### 1.2.4.2 Assessment of proliferation resistance (D132)

##### *Authors*

K.W. Hesketh (NNL)

##### *Description*

The European Union 6th Framework Plutonium MA (PUMA) project is investigating the potential of High Temperature Reactor (HTR) and Very High Temperature Reactor (VHTR) technologies as a possible means of burning plutonium and MAs. The fuel form for (V)HTR may either be prismatic blocks used in the General Atomics Gas Turbine Modular Helium Reactor (GT-MHR) or the pebble bed as used in the South African Pebble Bed Modular Reactor (PBMR). While the current generation of nuclear power plants are dominated by Light Water Reactors (LWRs), (V)HTRs potentially offer superior performance in term of efficiency and economy, inherent safety and possibly in proliferation resistance. If these advantages can be realised in practice, (V)HTRs could be competitive with LWRs for the future generations of reactor build (post-2020). The proliferation resistance assessment report [1] describes an assessment of the proliferation resistance performance of (V)HTR, with both prismatic and pebble bed type cores considered.

It considers several different proliferation pathways by which a state or sub-national organisation might attempt to acquire fissile or radioactive materials from the (V)HTR and its fuel cycle:

- *Pathway 1* postulates a proliferator diverting un-irradiated fuel for the purpose of constructing a fissile device.
- *Pathway 2* postulates a proliferator wanting to access fissile material in irradiated fuel.

- *Pathway 3* covers the potential for a proliferator to irradiate clandestine fertile targets to produce fissile material.
- *Pathway 4* considers the potential threat of a sub-national organisation stealing radioactive material for use in a dispersal weapon.

A systematic analysis of the different pathways is carried out to arrive at a qualitative comparison with LWR once-through and recycle scenarios and with a CANDU once-through cycle. This is complemented by a quantitative assessment made using a Multi Attribute Utility Analysis (MAUA). This is presented in the form of plots of the Utility Function  $U(\underline{x})$  versus the Access Function  $A(\underline{x})$ , the so-called U-A plots. The Utility Function is taken to be the product of the Value Function  $V(\underline{x})$  and the accessibility function  $U(\underline{x}) = A(\underline{x}) V(\underline{x})$ , where  $\underline{x}$  represents the multi-attribute vector comprising the various parameters of the nuclear system.  $A(\underline{x})$  is a function of the number of intrinsic barriers preventing access to the nuclear material.  $V(\underline{x})$  represents the intrinsic value of the nuclear material for the particular pathway concerned and is determined by its mass and isotopic composition. In this context it is preferable that  $U(\underline{x})$  should be as small as possible.

Pathway 1 postulates a proliferator diverting un-irradiated fuel for the purpose of constructing a fissile device. As the density of fissile material in (V)HTR fresh fuel is much lower than in LWR fuel assemblies, a proliferator would need to divert large volume of un-irradiated fuel to access a significant quantity of fissile material, increasing the risk of detection. The fissile material would also need to be separated from the graphite matrix and SiC shells and these extra processing steps can be regarded as additional physical barriers that are not present in LWRs. The qualitative assessment shows (V)HTR to be more proliferation resistant against Pathway 1 than current reactor systems, a conclusion which is confirmed by the quantitative MAUA analysis.

Pathway 2 postulates a proliferator wanting to access fissile material in irradiated fuel. (V)HTR has a significantly lower fissile inventory in irradiated fuel than current reactor systems and the fissile quality tends to be very poor (extremely so for uranium-free plutonium fuel). Combined with the reduced accessibility implied by the graphite matrix and the SiC shells, the qualitative ranking is strongly positive compared with current reactor systems. Again, the MAUA analysis confirms this result, as illustrated by the U-A plot shown in Figure 1. This shows a much lower Utility Function for both (V)HTR fuel cycle options considered relative to the values that apply LWR once-through and recycle (represented by EPR) and CANDU, which ranks (V)HTR highest for intrinsic proliferation resistance.

Pathway 3 covers the potential for a proliferator to irradiate clandestine fertile targets to produce fissile material. The qualitative assessment has identified that for the prismatic core (V)HTR there is very little scope for irradiating the tonne fertile masses needed to produce sufficient fissile material to pose a proliferation threat in a reasonable time. For the pebble bed (V)HTR there is possibly slightly more scope for loading fertile targets, but it would be difficult to irradiate a large fertile mass without detection. In both cases the need to separate the fissile material from the graphite matrix and SiC shells is beneficial in increasing the number of intrinsic barriers relative to conventional nuclear fuel designs. The quantitative assessment confirms the very high ranking of (V)HTR against this pathway.

Pathway 4 considers the potential threat of a sub-national organisation stealing radioactive material for use in a dispersal weapon. Various different scenarios have been explored for this pathway, including dispersal of plutonium, MAs from target fuels or irradiated fuel fragments. For dispersal of plutonium, the uranium fuelled (V)HTR significantly out ranks the other systems examined, because there is no separated plutonium in the fuel cycle and the plutonium inventory in irradiated fuel is low. However, the uranium-free plutonium fuelled

(V)HTR ranks roughly equivalent with EPR recycle, because separated plutonium exists at some part of the fuel cycle and this represents vulnerability. (V)HTR ranks better than current reactor systems as regards dispersal of irradiated fuel fragments, largely because the graphite matrix and SiC shells act as additional barriers.

#### *Conclusions & recommendations*

- *The qualitative assessment shows (V)HTR to be more proliferation resistant against Pathway 1 (diversion of un-irradiated fuel for the purpose of constructing a fissile device) than current reactor systems, a conclusion which is confirmed by the quantitative MAUA analysis.*
- *The qualitative ranking of (V)HTR is very favourable for Pathway 2 (undeclared access to fissile material in irradiated fuel for the purpose of constructing a fissile device) compared with current reactor systems. Again, the MAUA analysis confirms this result.*
- *The qualitative assessment for Pathway 3 (clandestine irradiation of fertile targets) has identified that for the prismatic core (V)HTR there is very little scope for irradiating the tonne fertile masses needed to present a proliferation threat. For the pebble bed (V)HTR there is possibly slightly more scope for loading fertile targets, but it would be difficult to irradiate a large fertile mass without detection. In both cases the need to separate the fissile material from the graphite matrix and SiC shells is beneficial in increasing the number of intrinsic barriers relative to conventional nuclear fuel designs. The quantitative assessment confirms the very high ranking of (V)HTR against this pathway.*
- *Various different scenarios have been explored for Pathway 4 (postulated threat of a sub-national organisation stealing radioactive material for use in a dispersal weapon), including dispersal of plutonium, MAs from target fuels or irradiated fuel fragments. For dispersal of plutonium, the uranium fuelled (V)HTR significantly out ranks the other systems examined, because there is no separated plutonium in the fuel cycle and the plutonium inventory in irradiated fuel is low. However, the uranium-free plutonium fuelled (V)HTR ranks roughly equivalent with EPR recycle, because separated plutonium exists at some part of the fuel cycle and this represents vulnerability. (V)HTR ranks better than current reactor systems as regards dispersal of irradiated fuel fragments, largely because the graphite matrix and SiC shells act as additional barriers.*
- *The study highlights that a prismatic (V)HTR core would be amenable to conventional safeguards accounting and verification procedures, with fuel blocks accounted for individually in the same way as LW fuel assemblies. However, a modified approach would be needed in pebble bed cores because of the impracticability of accounting for individual fuel spheres.*

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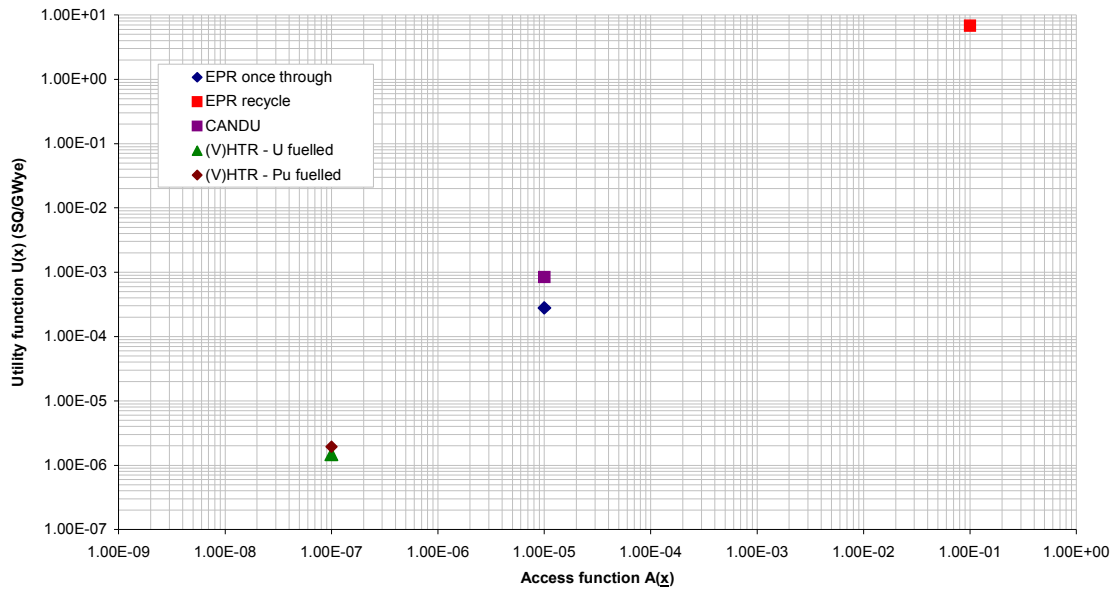


Figure 1: U-A plot for Pathway 2: Diversion of fissile material from irradiated fuels.



## 1.3 Work Package 2

### 1.3.1 Introduction

#### 1.3.1.1 Reminder of objectives of Work Package 2 (WP2)

This Work Package took a rational view of the requirements and design of innovative fuel forms for Pu and Minor Actinide incineration in High Temperature Reactors. The following aspects were covered with the design of an optimised coated particle fuel for irradiation testing in future more experimentally oriented programmes as a primary goal. Topics covered were:

- Establishment of helium behaviour in both Pu and MA kernels and PyC and SiC coating layers
- Development of helium and swelling behaviour models for coated Pu and MA particle fuels
- Design of coated particles suitable for ultra high burn-up of Pu and MA kernels
- Design of an irradiation facility meeting requirements for a future irradiation test in the HFR Petten

The participants were JRC-ITU (WPL), NRG, FZJ, TUD, GA, BN, NEXIA (now NNL). During the project BN ceased to operate, but their contribution was largely achieved prior to the cessation of their activities.

The deliverables and support documents from WP2 are listed below. Their output is summarised in this report.

No.	Title	Responsible
D211	PUMA kernel composition and coating geometries for WP1	JRC-ITU (Somers)
D212	Data review on past experience of Pu and MA fuels	BN (Tourey)
D213	Report on the development of a model for swelling behaviour and helium release in Pu/MA HTR fuels	BN (Tourey)
D214	Report on the modelling and design optimisation of Pu and MA based coated particles	NNL (Abram)
D215	Report on the coupling of core physics and CP design optimisation of Pu and MA based coated particles	TUD (Kloosterman)
D216	Chemical composition optimisation for Pu and MA kernels	NEXIA (Abram)
D221	Helium release kinetics from fuel kernels	JRC-ITU (Somers)
D222	Helium release from aged coated particles	JRC-ITU (Somers)
D231	Design and safety study for an irradiation vehicle for transuranic coated particles	NRG (Bakker)

S203	Fuel compositions and geometries of Pu/MA-based fuel	TUD (Jonnet, Kloosterman)
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### 1.3.1.2 General considerations for PUMA fuels in HTRs

The high temperature reactor (HTR) as means to dispose of long lived minor actinide waste is a medium term option, and so is potentially available before the introduction of a fleet of fast reactors (FR), either of current or Generation IV (Gen IV) designs. One strategy is outlined in Figure 2. Current day light water reactor (LWR) UO<sub>2</sub> fuel is reprocessed with some of the Pu returning to the LWR in the form of MOX. Remaining Pu not used for LWR MOX can be fabricated into HTR fuel. In addition, the Pu extracted from LWR-MOX spent fuel can also be fabricated in the form of HTR fuel. Pu and minor actinides from the LWR UO<sub>2</sub> and MOX fuels will be separated and be processed for treatment in the HTR. Irradiated HTR fuel can be considered for direct disposal or even reprocessing with the Pu and MA constituents being once again prepared for HTR irradiation. Other similar scenarios based on Gen IV fast reactors rather than LWRs can also be envisaged. In either case dedicated transmutation HTRs can be considered as a part of the nuclear park, where symbiosis with fast reactors is achieved.

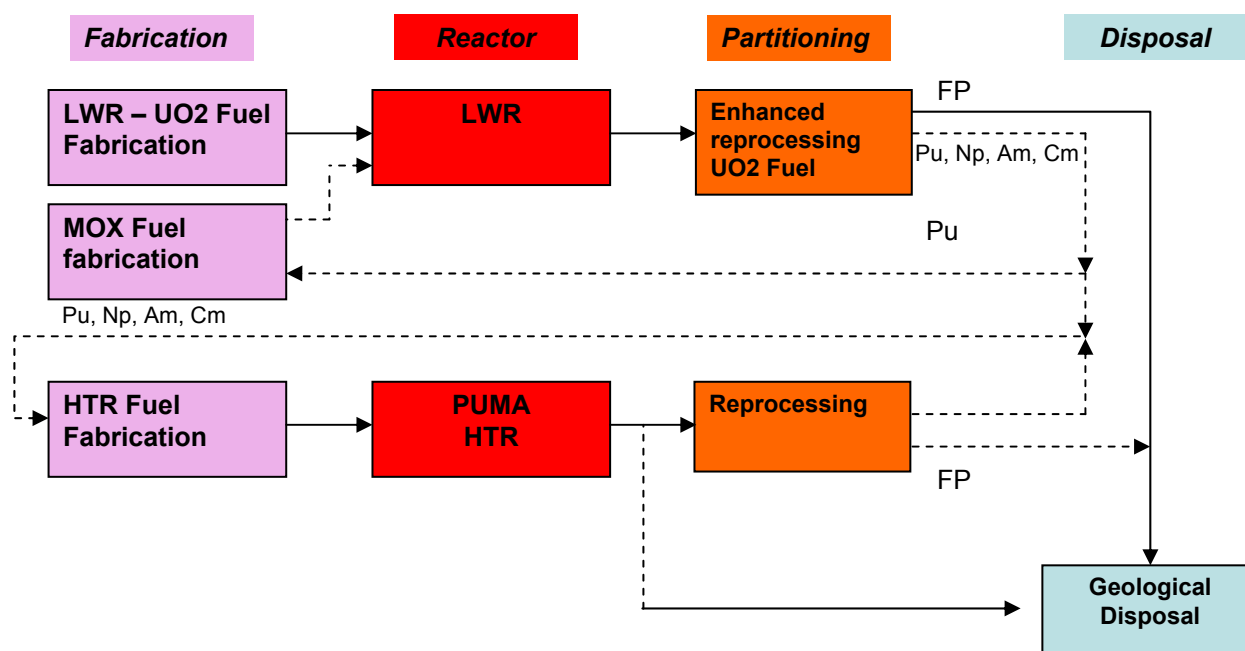


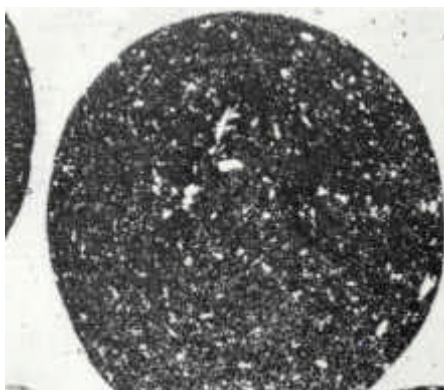
Figure 2: Transmutation Strategy with a PUMA HTR.

The HTR is highly flexible and can accommodate a full plutonium core. Pu based coated particles have been irradiated in the past and fuel burn-ups of 750 GWd/tonne were demonstrated in the experimental HTR Peach Bottom (USA) and Dragon (UK) test reactors. For transmutation purposes, ultra-high burn up plutonium (Pu) and Minor Actinides (MA)-based coated particle (CP) fuels will be needed. The steps required to meet these goals cover optimisation of the reactor core, making sure that the unique and unsurpassed safety features of the UO<sub>2</sub> fuelled HTR are maintained, and the development of novel fuel kernels capable of

either being reprocessed or suitable for direct geological disposal. The manufacture of HTR fuels incorporating Pu and/or minor actinides will be more difficult than for the corresponding pellet type fuels. There are advantages to be gained by the use of inert matrix based fuel kernel to dilute the fissile component.

### 1.3.1.3 Past investigations on Pu based coated particle fuel (D212)

The feasibility of Pu bearing particles was successfully demonstrated from the end of the 1960's by BELGONUCLEAIRE and SCK-CEN, who developed the technology for the manufacture and coating processes of HTR particles, and participated in several irradiation experiments. PuC fuel was the initial kernel composition, but was quickly transferred to pure (and more seldomly mixed) oxide. This was a necessary development as PuC has a significant vapour pressure, which resulted in unacceptable losses both during its production and during the coating steps. It is also noteworthy that at an early stage BN recognised the necessity of providing adequate voidage in the buffer layer so that highest burn-ups could be reached. Indeed BN introduced the "**diluted kernel**" concept – see Figure 3. In this way the kernel diameter is increased, which permits a larger free volume in the buffer layer, without increasing its thickness. BN used carbon as a diluent and used dilution factors between 5 and 30 (most commonly 20). Typical diluted kernel diameters were 500  $\mu\text{m}$ . Furthermore BN also found that use of an undiluted smaller pure  $\text{PuO}_2$  kernel resulted in poorer performance due to Pu migration through the buffer layer.



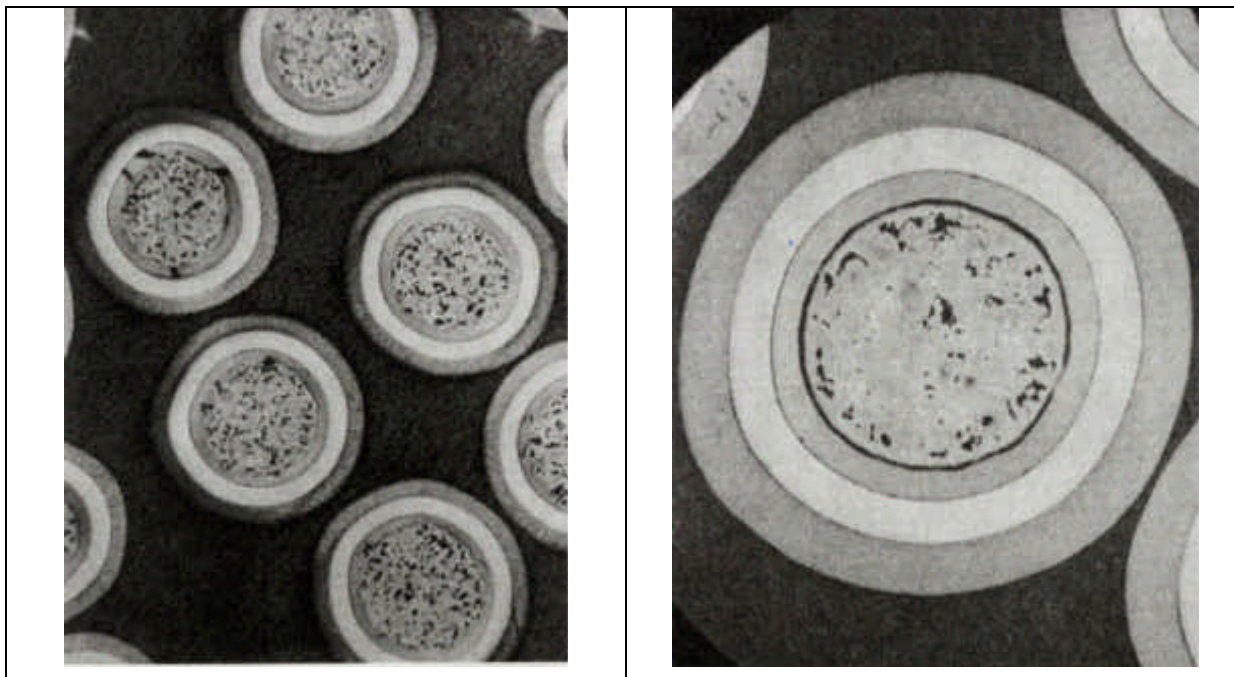
*Figure 3:  $\text{PuO}_2$ -20C kernel.*

The fabrication of the Pu kernel was based upon the agglomeration of  $\text{PuO}_2$  powder and carbon powder with furfuryl alcohol (10 cc FFA / 100g  $\text{PuO}_2$ -C). After mixing and granulation in a hand mortar, the  $\text{PuO}_2$ -C green crumbs were sieved to the correct size range  $\pm 100\mu\text{m}$  from the mean size and then spheroidized in a planetary mill. The kernels were then hardened by polymerisation of the binder FFA in acid vapour for 24 hours. The resin-bonded kernels were heated in order to decompose the furfuryl alcohol polymer and to reduce gas evolution during the coating. To avoid plutonium volatilization by reaction of plutonium oxide with carbide, a carbon monoxide atmosphere (1 atm.) was applied during 2 hours up to 1800°C. The classical sol gel routes as applied for  $\text{UO}_2$  fuels were applied by other laboratories (ORNL, CNEN, KEMA and BN). In all cases the Pu oxidation state was reduced from IV, to hinder CO production during irradiation Typical O/M ratios lay between 1.7 and 1.9.

The coating of these particles was made using standard methods for Buffer, iPyC, SiC and oPyC layers. Given that there is significant friction in the fluidised bed of the chemical vapour deposition (CVD) furnaces, Pu contamination on the external surfaces of the coated particles was observed (10kBq/gPu in the kernel. Two to three washing steps using water-15% acetone mixtures were sufficient to decrease the surface contamination by two orders of magnitude.

Irradiation testing was made in several reactors under a variety of conditions. In the first of these coated particles with PuO<sub>2</sub>-20C were irradiated in the R2 reactor at Studsvik to burn-ups between 20 and 37%FIMA. At 1450°C (already a high temperature for coated particle fuel), release to birth (R/B) ratios for Xe were about  $5 \times 10^{-6}$ . At very extreme temperatures (1850°C) this value rose to  $10^{-2}$ . In neither case was Pu release detected.

A second more realistic test was performed on loose coated particles (at 1300°C) in the DRAGON reactor, during an irradiation for 224 effective full power days (EFPD). Up to 90% FIMA was reached and the fast neutron flux was  $1.8 \times 10^{21}$  n.cm<sup>-2</sup> (>0.1MeV). Again an excellent behaviour was found (see Figure 4).



*Figure 4: DRAGON experiment – PuO<sub>2</sub>-20C (BU = 90% FIMA).*

A temperature of about 1300°C seemed to be the limit for satisfactory operation of the Pu coated particle fuels. Above this temperature, the integrity of the particles was not preserved anymore. The R&D on HTR fuel was wound down in Belgium from 1975, with the dismantling of the uranium and plutonium fabrication lines, the shut-down of the DRAGON reactor and the end of the DRAGON project collaboration.

The results obtained in these early investigations indicate clearly that PuO<sub>2</sub> and eventually MAO<sub>2</sub> fuels should behave correctly, if appropriate irradiation conditions are chosen.

## 1.3.2 Coated Particle fuel: geometry and composition

### 1.3.2.1 Coated particle geometry (D211)

The traditional TRISO layers used in U based fuel are taken for PUMA fuels. Their characteristics are given in Table 1.

Table 1: Coated particle layers.

Layer	Layer thickness (µm)	Layer Density (g.cm <sup>-3</sup> )
Buffer	90	1.05
iPyC	40	1.9
SiC	35	3.18
oPyC	40	1.9

Concerning the fuel kernel diameter, the ultimate design will come from core physics investigations. Nevertheless, it is certain that the pure PuO<sub>2</sub> kernel will be very much smaller than the 500 µm considered today for UO<sub>2</sub> fuel. This is simply due to the high fissile component as compared to low enriched uranium. PuO<sub>2</sub> kernel diameters of 200 µm are likely.

Within the PUMA project, separation of U/Pu is considered along with complete minor actinide (MA) separation or partial separation. This give rise to three possible fuel types:

- (a) PuO<sub>2</sub> (probably with NpO<sub>2</sub>)
- (b) MAO<sub>2</sub>
- (c) (Pu,MA)O<sub>2</sub>, whereby the Pu:MA ratio is significantly lower than in spent fuel.

Their loading in the reactor should be based on heterogeneous compacts (pebbles or blocks) for each. This is again to separate the difficulties in fabrication. Such a heterogeneous loading is not practical however, as it would lead to higher operating temperatures of the CPs in the Pu fuelled compacts. Nevertheless individual compacts can be formed from a mixture of heterogeneous Pu and MA based coated particles so that the temperature in the CPs is more "uniform" throughout the core. Recycling of Cm is considered as not practical.

One real advantage of the HTR is its low power density of the core. This results in an abundance of space, so that fuel designs are more open than for other reactor systems. The early studies of Belgonucleaire clearly identified the advantages in diluting the Pu in carbon. Other promising materials are given in Table 2.

Table 2: Diluting inert matrices for HTR actinide (An) fuel.

	Matrix	Fuel/target
YSZ	$Zr_{0.85}Y_{0.15}O_2$	$(Zr_{0.85}Y_{0.15})_{1-z}An_zO_2$
YSZ-Ce	$(Zr_{0.85}Y_{0.15})_{1-y}Ce_yO_2$	$((Zr_{0.85}Y_{0.15})_{1-y}Ce_y)_{1-z}An_zO_2$
Ceria	$CeO_2$	$Ce_{1-z}An_zO_2$
PYR (pyrochlore)	$Zr_2Ce_2O_7$	$(Zr_2Ce_{2-z}An_z)O_7$
Graphite	C	C + $AnO_2$

Yttria stabilised zirconia (YSZ) has been the subject of a number of studies as an inert matrix, It provides an ideal option for fuels to be sent directly for geological disposal. As a consequence, it is difficult to reprocess. Ceria has an added chemical advantage, as it can be incorporated in the fuel in its trivalent (III) form so that oxygen released through fission of the actinides can be gettered via the Ce(III/IV) buffer. Reprocessing of carbon based kernels would then be reduced to pyrolysis of the carbon in a head end step.

The premise in designing the diluted kernel relies on the mass of the fissile component being the same as the pure 200  $\mu\text{m}$  actinide oxide kernel. Staying with tradition, justified by the excellent results obtained with NUKEM produced  $UO_2$  fuel, the diluted kernel diameter should be 500  $\mu\text{m}$ . At this size, one can expect the in pile fuel performance i.e. that of the sealing coating layers, to be similar to that of a  $UO_2$  kernel, operated at the same power/kernel.

### 1.3.2.2 Chemical Composition Optimisation for Pu and MA kernels (D216)

One aspect of the development of the fuel design for High Temperature Reactors is the assessment of the chemical behaviour of an optimised coated Pu and minor actinides (MAs) particle fuel for irradiation testing to high burn-up. The objective of such a study is to provide information that could assist in the selection of recommended fuel kernel compositions for use in experimental programmes in the future. Deliverable D216 of PUMA Project Work Package 2 comprised the evaluation of the chemical processes contributing to the performance of fuel particles containing MAs and the collation of experimental data on the variation of oxygen potential with fuel composition.

A thermodynamic model and database was developed for oxide fuel compositions considered by the PUMA Project and valid over the temperature range 1000-1600°C. This included the Gibbs energy variation with temperature and composition for the pure substances and solution phases which are represented as either ideal or non-ideal. Calculations using the database were performed to predict the evolution of the oxygen potential of the fuel and gas pressure during irradiation and evaluate the influence of ceria as an oxygen getter on the chemical behaviour. The thermodynamic model included pure substance and solution data for the oxide components  $UO_2$ ,  $NpO_2$ ,  $PuO_2$ ,  $AmO_2$ ,  $CmO_2$  and  $CeO_2$  and was validated against experimental oxygen potential results. Whereas the data for the uranium, plutonium and cerium systems were extensive, information for the americium and neptunium systems were limited and non-existent for the curium system. Although the latter constituent was not included in the PUMA reference compositions, further experimental data on the americium and neptunium systems are required to validate the model over a wider range of actinide

content and O/M compositions. Scoping calculations for the binary and ternary systems were performed using the model to assess the influence of the components of the particle fuel on the overall oxygen potential of the system. The main contributor to the increase in oxygen potential of the system was americium.

For the PUMA reference compositions,  $(\text{Pu,Np,Am})\text{O}_{2-x}$ , an increase in the oxygen potential was determined for the substitution of plutonium by MAs, in particular americium. This increase was more significant for O/M ratios above  $\sim 1.90$  and increased with americium content. The effect of the addition of cerium as a diluent depended on the initial O/M ratio and the cerium and actinide concentrations. The variations of oxygen potential with O/M ratio for the two reference fuels (Pu-O and MA0%Ce) and ceria diluted composition (MA20%Ce) are shown in Figures 5 (a) and (b).

Calculations were also carried out to determine the effect of fuel composition and temperature on the gas pressure generation in the particle fuel-carbon system during irradiation. Contributions from the release of the volatile fission products and helium were also included in the assessment. An increase in the initial Am concentration resulted in an increase in the oxygen potential, a decrease in the final O/M ratio and hence an increase in the gas pressure. This pressure was reduced by lowering the initial O/M ratio of the oxide. The effect of introducing cerium as an oxygen getter resulted in a slight increase in the oxygen potential and gas pressure for an initial O/M ratio of 2.00. However, a reduction of the gas pressure was determined for initial O/M ratios less than  $\sim 1.90$ . The cerium oxide would need to be added to form a single solid solution phase with the actinide oxides and the O/M ratio adjusted to the appropriate value.

The main conclusions from this study were that the evolution of the chemical constitution with burn-up for the PUMA fuel compositions should not result in excessive internal pressurisation provided the concentration of americium is low and the initial O/M ratio reduced. The effects are also minimised by the addition of an appropriate amount of cerium in solid solution. These conclusions are only valid for total concentrations of Am and Np less than  $\sim 0.3$  and for irradiations up to 15% FIMA. The influence of these chemical modifications on the amoeba effect is uncertain particularly if solid state diffusion is the dominant mechanism rather than gas phase transport. Since both processes are dependent on the oxygen potential of the system and the changes are not predicted to be too significant, the incidences of kernel migration in actinide oxide fuel particles might not increase compared to conventional HTR oxide fuel.

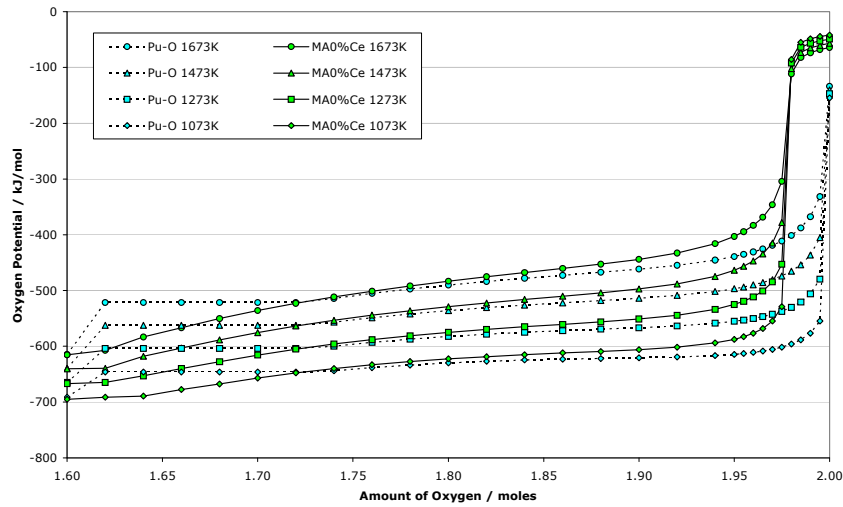
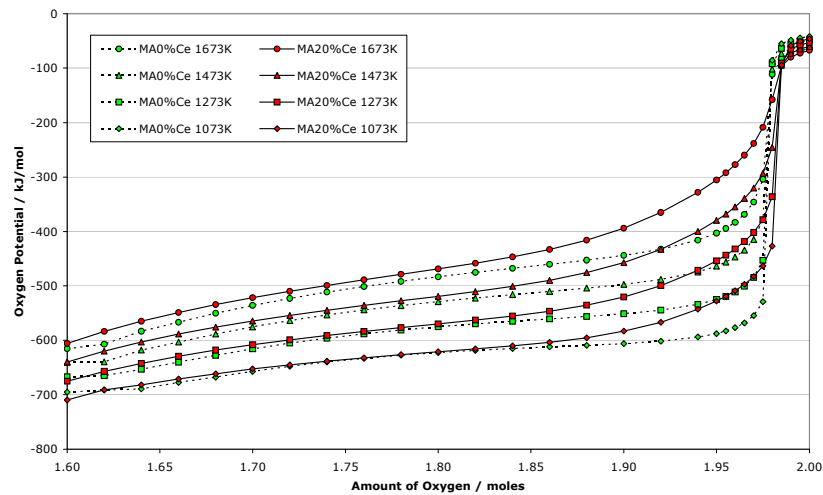

 (a)  $\text{PuO}_{2-x}$  and  $\text{Pu}_{0.889}\text{Am}_{0.042}\text{Np}_{0.069}\text{O}_{2-x}$ 

 (b)  $\text{Pu}_{0.889}\text{Am}_{0.042}\text{Np}_{0.069}\text{O}_{2-x}$  and  $\text{Pu}_{0.712}\text{Am}_{0.033}\text{Np}_{0.055}\text{Ce}_{0.2}\text{O}_{2-x}$ 

Figure 5: Calculated oxygen potentials for PUMA reference fuel compositions.

### 1.3.3 Helium release from kernels and coated particles

#### 1.3.3.1 Helium release from kernels (D221)

As the HTR fuel operates at a temperature around 1200°C, the He-release fraction at this temperature is decisive for the modelling. PUMA is concerned with Pu and minor actinide (MA) kernels, for which there are no representative samples available, on which measurements can be made directly. In the absence of data direct data, recourse is made to release data from a variety of other sources. These include

- Irradiated  $\text{UO}_2$  fuel
- Irradiated MOX fuel
- Irradiated  $\text{MgAl}_2\text{O}_4$  –  $\text{AmAlO}_3$



For spent fuel the majority of the helium is produced after the irradiation itself, indeed when the fuel is in storage. Thus the mechanisms driving its release may well be different from those prevalent in the reactor, e.g. temperature, which in pellet fuel varies greatly across the pellet diameter, with the highest temperatures, similar to those encountered in HTRs, found at the central axis.

In the HTR, the temperature gradient in the kernel is small, i.e. less than 20°C. The operating conditions are reactor dependent and are summarised in Table 3 for pebble bed fuel reactor systems. The fractional helium release from fuel at a temperature corresponding to the upper limit for VHTR operation, i.e. 1100-1200°C, is most important

*Table 3: HTR UO<sub>2</sub> Fuel operating temperatures.*

Parameter	HTR MODUL	HTR 10	PBMR	HFR-EU1bis*
Max temp. (°C) Gas outlet	750	750/900	900	
Max temp. (°C) fuel surface	926	831	1000	1050
Max temp. (°C) fuel centre	1130	864	1100	1250

\* HFR EU1bis is an irradiation test in the HFR Petten

Helium release measurements are best carried out in a Knudsen-cell facility. The fuel samples examined here were irradiated at low temperature up to a high local burn-up (95-100 GWd/t). The largest fraction of helium was created by decay of actinides and was dynamically dissolved in the lattice during post-irradiation storage. Figure 6 shows the helium release measurements and that of <sup>136</sup>Xe. In a further study the He and Xe release from a dedicated Am transmutation target (MgAl<sub>2</sub>O<sub>4</sub> – AlAmO<sub>3</sub>), irradiated in the HFR Petten has been measured. The helium and xenon release both begin at about a 1000°C. While the helium is released at about 1300°C, only about half of the Xenon is released at this temperature.

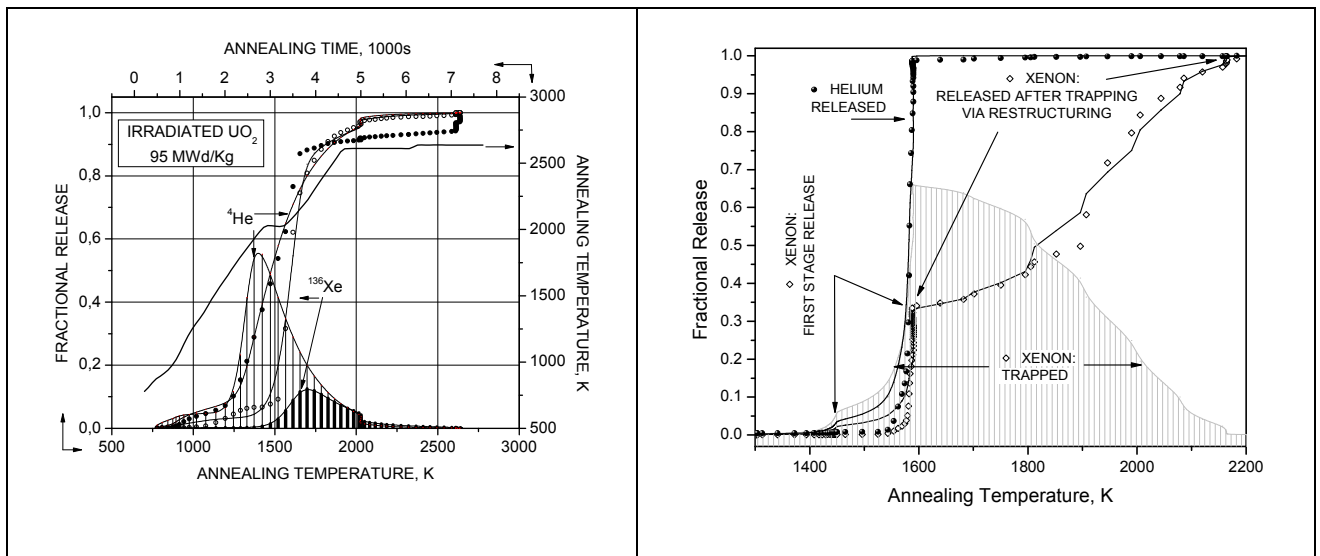


Figure 6: He and Xe release from irradiated  $\text{UO}_2$  (left) and a dedicated Am transmutation target  $\text{MgAl}_2\text{O}_4\text{-AmO}_2$  (right).

These results from these different samples indicate that major helium release occurs at typical HTR fuel operating temperatures. The measurements described here are all based on dynamic studies in which the temperature of the sample is ramped. The heating rate influences the derived release temperature in a minor way, and usually induces a higher release temperature for higher heating rates. In the reactor however, the fuel is operating under isothermal conditions, and the helium will be released continuously. Helium release studies on other ceramics including zirconolites and pyrochlores with fluorite based structures have been performed. The samples were doped with strong alpha-emitters ( $^{238}\text{Pu}$ ,  $^{244}\text{Cm}$ ) in order to generate high amount of helium. The onset for helium release was even lower for these compounds and started at around 600 K.

**In conclusion, it is recommended that full helium release from the fuel kernel should be considered for modelling purposes.**

### 1.3.3.2 Helium release from coated particles (D222)

The ability of a coated fuel particle (CP) to achieve high burn-up is determined by the stability of the particle layers against irradiation effects, and the build up of gas pressure inside the CP. In contrast to  $\text{UO}_2$  fuels, Pu and MA fuels will generate significant quantities of helium which, depending on the fuel composition, can be far greater than the fission gas. As the HTR fuel kernel operates at temperatures above 1500 K, helium release from the kernel to the highly porous buffer layer will be complete, so that it acts as an additional pressure source during irradiation.

Helium release from coated particles in the PUMA programme constitutes the only new experimental results. Coated particles were supplied by Belgonucleaire were composed of kernels  $\text{PuO}_2$  and the coatings were in the standard TRISO format. The coated particles had been manufactured in the late 1960's, thus permitting ample time for radiogenic helium to be generated from decay of Am (a decay product of  $^{241}\text{Pu}$ ). Thus, these results pertain to fresh

fuel and it is not clear that the results will be similar for irradiated material, and even then, this could be a function of burn-up, due to the damage that is caused in the carbon and SiC coatings.

**No helium release could be observed even at the highest temperature (2800 K).** In addition, the constituents of the different coating layer barriers (C, SiC) did not show a pronounced release before the (smooth) onset of their vaporization at very high temperature (2400 K), as can be observed in Figure 7. **Evaporation of plutonium species from the fuel kernel was not observed at all.** The SEM micrograph in Figure 8, recorded after heating at 2800 K, shows no evidence of rupture or major morphological modification during the annealing process.

These results confirm that the helium remains confined within the coated particle. Most likely, as observed in former experiments, the helium is released from the plutonium oxide kernel but then is retained in the porous graphite buffer layer by the tight iPyC/SiC/oPyC sealing layers.

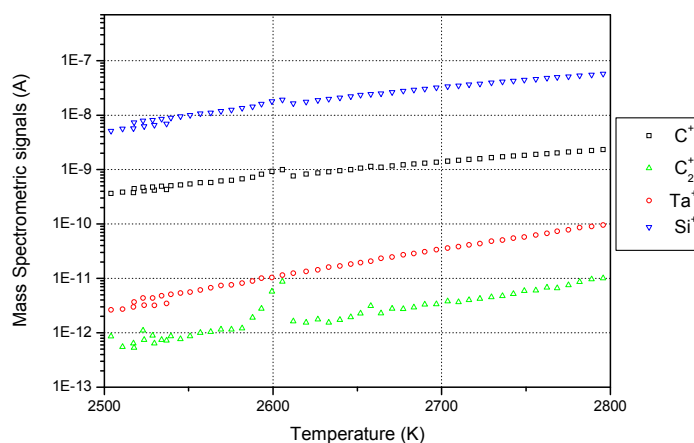


Figure 7: Knudsen cell mass-spectrometer signal for Si, Ta and graphite (C and C<sub>2</sub>). The Tantalum from the TaC crucible is also seen in the vaporization curves. No helium or Pu release was detected.

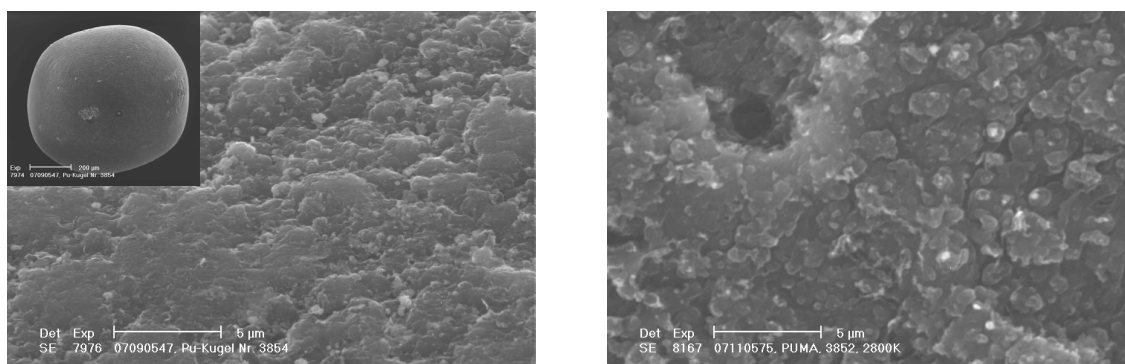


Figure 8: Scanning electron micrographs of a coated particle (inset top left) and higher magnification view of the graphite surface before (top) and after annealing at 2800 K (bottom).

A second control measurement was made to verify that the particle had not been damaged and in fact lost all its helium prior to the Knudsen cell test. In this second control experiment, the coating layers were deliberately destroyed during annealing and both helium and Pu could be detected.

These un-irradiated coated particles exhibit remarkable thermal and mechanical stability and excellent helium retention capacity at temperatures higher than 1000 degrees above the normal operating temperature of this type of fuel. For modelling of the fuel behaviour, these results indicate that the **full helium retention** should be included in the model for helium pressure build up (in normal operating conditions and also in cases where loss of coolant flow and pressure might occur).

### 1.3.4 Performance Modelling (D213, D214, D215)

#### 1.3.4.1 Swelling and Helium models

Thermo-mechanical properties of interest of the fuel kernel are those for the isothermal and steady state thermal conditions. They can be subdivided into instantaneous properties, such as:

- Thermal expansion. For mixed  $\text{UO}_2$  and  $\text{PuO}_2$  the thermal expansion of the solid is found by combining the contribution from each constituent in proportion to its weight fraction.
- Elastic modulus, or Young's modulus, which is affected by the temperature, porosity and the plutonium density in the fuel.
- Poisson modulus.
- Theoretical density as a function of fuel composition and stoichiometry.
- Kernel swelling. This effect is modelled by different mechanisms, like solid fission product matrix swelling, pseudo-solid fission product swelling (e.g. caesium), densification or in-pile shrinkage, fuel swelling accommodation, and fission gas bubble swelling.

In addition, long-term effects, which require time or burn-up/fluence to proceed, should be included. Examples are:

- Swelling, densification and accommodation
- Fission gas release
- Creep and plasticity
- CO generation and structural changes and other effects

The fission gas release model in COCONUT is a deterministic "best estimate" diffusion model derived from Booth's model. It has been extensively tested on  $\text{UO}_2$  fuel in steady state and transient operation. In the model it is assumed that the gas atoms diffuse in the (equivalent) sphere; that the boundaries of the sphere behave as a perfect sink; that the gas atoms trapped are motionless, and that the probability of capture of gas is independent of the gas concentration in solution.

In the CRYSTAL code the diffusion equation is solved numerically without approximation. Although demanding in terms of CPU time, it can deal with non-zero boundary conditions. For quick calculations, it also has an option to solve Booth's model extended to cyclic irradiation

histories (simulating pebble flow through the core). In PANAMA the fission gas release is calculated applying Booth's model, using a diffusion coefficient that can be adapted to the fuel type considered.

Helium diffuses far better in UO<sub>2</sub> than xenon and krypton. Consequently, the fractional helium release is much larger than that the fractional fission gas release. In COCONUT an empirical relation is used for the helium release fraction based on measurements with MOX fuel rods, while in the other codes (CRYSTAL, PANAMA and STRESS3) it is assumed that all produced helium diffuses directly out of the kernel.

CO production in all four codes is accounted for by an empirical relation based on (some version of) Arrhenius law describing the temperature dependence of a chemical reaction.

#### 1.3.4.2 Fuel performance and neutronics coupling (D215)

An important aspect in proposing alternative fuel compositions for transmutation purposes in High Temperature Reactors is to ensure that the Pu and MA-based coated particles show similar behaviour during irradiation as the U-based particles. For this purpose, the coupling between core physics and fuel performance codes is required. This allows the calculation of TRISO particle failure fraction based on realistic irradiation conditions. The analysis was performed for the pebble-bed reactor core geometry, although calculations for other geometries are possible as well. Furthermore the analysis was limited to steady-state operating conditions.

A recent core physics code system was extended for the calculation of the equilibrium composition of Pu and PuMA-loaded pebble-bed HTR cores. This code system makes use of existing and well documented codes or modules (SCALE-5, THERMIX) and of a neutron diffusion code (DALTON) that proved to provide good results in benchmark exercises. For the fuel performance calculations, the particle stress analysis code CRYSTAL has been used. To this end, CRYSTAL has been extended with BAF-dependent thermal expansion of the coating layers, with the contribution of Helium in the buffer pressure build-up (100% release is assumed), with PyC layer cracking and the effects of that on the SiC layer, with a fast and accurate fission gas release model, and with Monte Carlo sampling of particle position, geometry and material properties.

Application of the coupled core physics and fuel performance codes to two PUMA reference fuels has been performed:

- PuO<sub>1.7</sub> (first generation Plutonium) TRISO particles with a 200 µm diameter kernel, referred as the 1<sup>st</sup> reference fuel.
- (Y,Zr,An)O<sub>1.7</sub> (An taken from recycled PWR spent fuel after five years of cooling) TRISO particles with a 500 µm diameter kernel, referred as the advanced reference fuel. The molecular formula was chosen to be:



For both reference fuels, the thicknesses of the buffer, inner PyC, SiC and outer PyC coating layers were chosen to be 90/40/35/40 µm as a starting point.

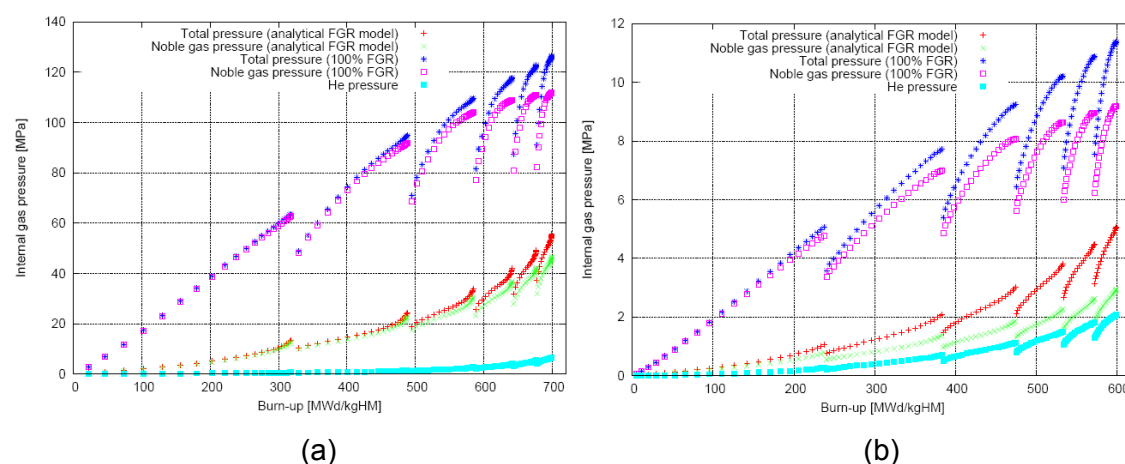
The equilibrium fuel composition and fluxes have been calculated and are in reasonably good agreement with results from other partners. The performance of the fuel has been assessed through the calculation of the TRISO particle failure fraction during irradiation, with particular attention to the effects of fission gas release and pyrocarbon failure on the mechanical

behaviour of the layers. The same calculations were performed with a U-loaded core for comparison. In Table 4 it is shown that the 1<sup>st</sup> reference fuel particle is not a good design, due to a very high failure fraction even if the pyrocarbon layers are assumed not to fail during irradiation. On the contrary, the advanced reference fuel particle shows a much better performance. Although the calculated failure fractions are not verified against experimental results, the trend shows that the (Y,Zr,An)O<sub>1.7</sub> TRISO fuel can be considered as a good option for both the transmutation capability and for the performance during irradiation.

**Table 4: Calculated failure fractions of UO<sub>2</sub>, PuO<sub>1.7</sub> and (Y,Zr,An)O<sub>1.7</sub> fuel particles for different configurations of fission gas release and stress models.**

Case	FGR	PyC failure	UO <sub>2</sub>	PuO <sub>1.7</sub>	(Y,Zr,An)O <sub>1.7</sub>
1	analy. 5	yes	$6.52 \times 10^{-5}$	$4.61 \times 10^{-2}$	$5.50 \times 10^{-6}$
2	analy. 5	no	$< 10^{-7}$	$3.056 \times 10^{-4}$	$< 10^{-7}$
3	approx	yes	$3.54 \times 10^{-5}$	$2.31 \times 10^{-2}$	$2.80 \times 10^{-6}$
4	approx	no	$< 10^{-7}$	$1.33 \times 10^{-5}$	$< 10^{-7}$
5	100%	yes	$7.519 \times 10^{-4}$	$2.16 \times 10^{-1}$	$2.83 \times 10^{-5}$
6	100%	no	$< 10^{-7}$	$9.095 \times 10^{-4}$	$< 10^{-7}$

The difference in the calculated failure fractions between the two PUMA reference fuels comes mainly from the buffer pressure during irradiation. In the case of the advanced reference fuel, the dilution ratio of minor actinides into the Ytria-stabilised Zirconia matrix (Eq.1), and the kernel diameter were chosen to enlarge the buffer layer such that it can accommodate approximately the same amount of gaseous fission products but more Helium than those of the 1<sup>st</sup> reference fuel design. The failure fractions presented in Table 4 and the buffer pressures in Figure 9 (a) and (b) show that this aim has been achieved.



**Figure 9: Contribution of Noble Gas and Helium in the total buffer pressure as a function of burn-up, calculated with the analytical FGR model or assuming instantaneous release (100%) for the (a) 1<sup>st</sup> and (b) advanced reference fuels, respectively.**

Although many improvements are still possible for both the core physics and the stress analysis codes, the present results show that the design and performance of advanced fuels during irradiation can be analyzed.

### 1.3.4.3 Irradiation performance modelling

The mechanical performance of the following coated particle fuels were analysed under 400 MWth PBMR-like irradiation conditions. The irradiation temperature, however, was kept constant at 1000°C:

- #1. 500 µm UO<sub>2</sub> reference particle, up to 96 MWd/kg HM or 10.4% FIMA.
- #2. 200 µm UC<sub>2</sub> particle, up to 700 MWd/kg HM or 78% FIMA.
- #3. 200 µm PuO<sub>1.7</sub> particle, up to 700 MWd/kg HM or 78% FIMA.
- #4. 200 µm (Pu,MA)O<sub>1.7</sub> particle, up to 600 MWd/kg HM or 66% FIMA.
- #5. 500 µm Ytria stabilised zirconia (YSZ) diluted (Pu,MA)O<sub>1.7</sub> particle, up to 600 MWd/kg HM or 66% FIMA.

Particle #1 is the German, South African and Chinese reference particle and is expected to survive this irradiation in good condition. Particle #2 is the reference fissile particle of the old US programme and is a borderline case under modern performance requirements. Particle #3 is the reference fissile particle of the Russian-American GT-MHR programme with good mechanical performance.

PuMA particles #4 and #5 have the composition (Pu,MA)O<sub>1.7</sub> and contain an identical amount of heavy metal and fissile material per particle. The 200 µm kernel diameter of particle #4 shows poor performance in the mechanical failure prediction. This, in part, is due to the lack of knowledge of fission gas and He retention, where conservatively 100% release had been assumed. The situation is much easier for the 500 µm kernel diameter (Pu,MA)O<sub>1.7</sub> particle with 15 % dilution in YSZ dilution where enough buffer layer is provided to avoid particle failure even under very conservative assumptions. In summary, predicted failure levels are given in Table 5.

*Table 5: Assessment of coated fuel particle performance for PUMA fuels.*

	#1 500 µm UO <sub>2</sub>	#2 200 µm UC <sub>2</sub>	#3 200 µm PuO <sub>1.7</sub>	#4 200 µm (Pu,MA)O <sub>1.7</sub>	#5 500 µm (Pu,MA)O <sub>1.7</sub> with 15YSZ
End-of-Life Failure Fraction	$3 \times 10^{-5}$	$6 \times 10^{-5}$	$2 \times 10^{-5}$	$9 \times 10^{-5}$	$1 \times 10^{-6}$
Acceptable	YES	NO	YES	NO	YES

on the basis of a limit of  $3 \times 10^{-5}$  failure fraction in mechanical performance predictions.

There are a number of drawbacks in these studies, especially as they concentrate on normal operation of the fuel. Transients really need consideration also. Kernel swelling is not an

issue as a large sacrificial buffer layer is provided to act as free space for the gases. Also, the kernel mechanics and coating mechanics are completely decoupled.

A small dense (Pu,MA)O<sub>1.7</sub> particle cannot survive irradiation with current coating materials and designs. A YSZ-diluted (Pu,MA)O<sub>1.7</sub> particle has excellent chances for survival.

### 1.3.5 Irradiation of actinide bearing coated particle fuels (D231)

For the development of plutonium/minor actinides based HTR fuel, irradiation testing of such fuel in a materials testing reactor (MTR) will be needed. Currently JRC-ITU in Karlsruhe is preparing the fabrication of coated particles containing plutonium/minor actinides. When fabricated, an irradiation test on loose coated particles and eventually on coated particles incorporated into spherical or cylindrical compacts is essential to qualify the fuel. Testing of loose coated particles is essential for the development of the final fuel form (i.e. compacts). It should be ascertained that the quality of these loose coated particles is sufficient.

In the past loose coated particles containing amongst other ThO<sub>2</sub>, (Th,U)O<sub>2</sub>, UO<sub>2</sub> and UCO have been irradiated. Aspects complicating the proposed irradiation of minor actinides coated particles in comparison with Th or U based fuels are:

- The gamma radiation coming from the minor actinides is stronger than the radiation coming from the thorium and the uranium. Thus it requires adequate protection measures and the introduction of non trivial handling procedures
- The surface of the minor actinides containing particles could be contaminated, though washing procedures could be used to reduce surface contamination to that which is permanently fixed on the particle.
- Irradiation experiments of minor actinides containing materials require specific safety precautions.

The design of the holders in which loose coated particles can be irradiated needs careful evaluation, adapting previous designs so that these goals can be achieved. Two options transpire:

- Irradiation of a loose bed of particles
- Irradiation of the particles with each particle having its own well defined location with direct contact with the holder in which the coated particles are placed.

Following these considerations the folder design in Figure 10 has been proposed.



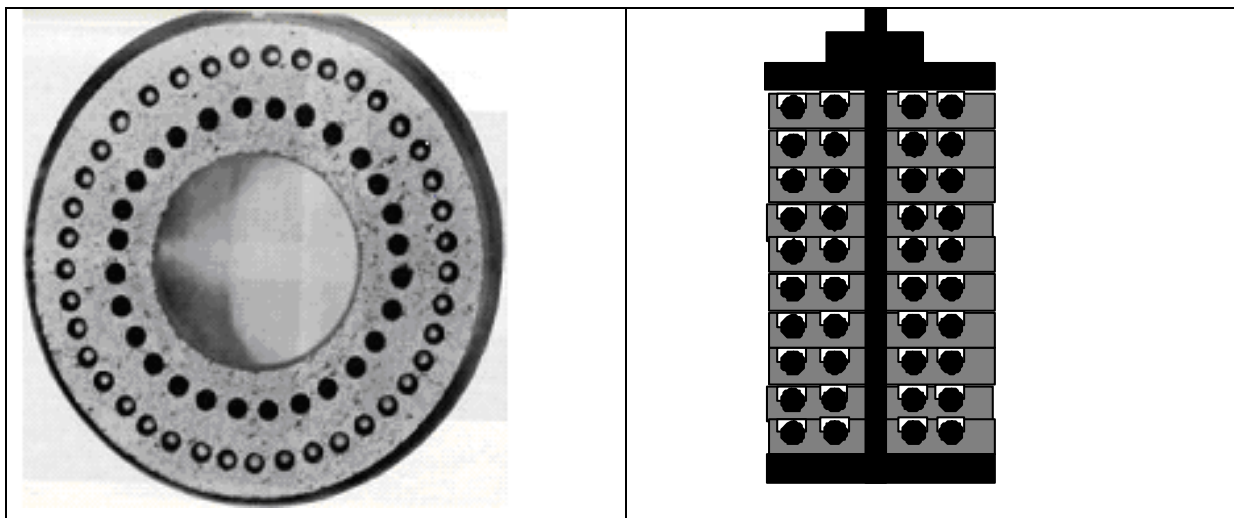


Figure 10: Holder design for the irradiation of loose coated particles.

The technical feasibility of these options has been assessed within the technical boundary conditions of the HFR Petten. Four of the capsules described in Figure 10 can be mounted in a rig spanning the core of the reactor. The flux buckling of the core can then be used to generate two high power/high burn-up capsules and two lower power/lower burn-up capsules. The gas gaps will be tuned in such a manner that there are two higher temperature capsules (CP temperature = 1100 °C) and two lower temperature capsules (CP temperature = 800 °C). The irradiation time will be sufficiently long to achieve fission of 50% of the actinides present, which will cause almost all <sup>241</sup>Am to be either fissioned or transmuted into curium or plutonium.

Monitoring of the experiment must also be considered. Positioning of the coated particles in individual well defined dimples ensures that they can be recognised and recovered at the end of life. Though gas purge systems are extremely useful as the onset of particle failure can be detected, they will not be considered to limit cost, and avoid handling difficulties that would necessarily ensue should minor actinides be present in the fuel. With these requirements An irradiation can be considered within a TRIO facility at the C3/C7 position in the HFR Petten. For an irradiation time of 560 EFPD a 30% actinide burn-up can be achieved with thermal and fast ( $E > 0.1$  MeV) fluences of  $5 \times 10^{25} \text{ m}^{-2}$  and  $2 \times 10^{26} \text{ m}^{-2}$  respectively.

### 1.3.6 Conclusions

Within this PUMA project a number of important results have been reported. When dealing with minor actinide bearing fuel helium generation is an important issue. Experiments have shown that He will be released from the kernel, but not from fresh kernels. Indeed, fresh fuel has shown a remarkable stability – up to 2500°C. For modelling purposes, 100% release of helium from the kernel is justified. The diluted kernel concept was first invoked by Belgonucleaire brings many benefits. The modelling studies have clearly indicated the advantages that can be gained by dilution. Essentially, for a given buffer layer thickness, more volume is available to accommodate the CO and He released. Chemical thermodynamic models have been deployed to design a kernel that will show limited CO production. The most important point here is that substoichiometric Pu or Am oxides are essential. Further improvement can be achieved by chemical buffering of the fuel by the addition of Ce sesquioxide, which takes up oxygen to form the dioxide. Ultimately any coated particle design must be validated in an irradiation test. Though not possible to perform an irradiation

programme in the PUMA project, the feasibility of such a programme has been demonstrated, and the initial data needed to launch such a test has been generated.

### 1.3.7 Recommendations

An irradiation test of coated particle fuel is essential for its qualification. The design studies made in PUMA have shown that the yttria stabilised zirconia coated particle fuel is an optimal choice. Thus future programmes must consider the fabrication, irradiation, post irradiation examination (PIE) and safety testing as a means to validate the fuel. Furthermore, it has been assumed that helium generated is not released from the coated particle. An irradiation test should also be performed to validate this hypothesis.

The largest drawback of the PUMA studies is the concentration on mechanical performance during normal operating conditions. Equally, and perhaps more, important are aspects of fission product transport and accident (transient) behaviour. This necessitates studies of Ag and Pd release, which will be more important from Pu fuels. The resulting SiC corrosion will need more experimental work. New measurements of SiC strength on modern TRISO particles need to be made and incorporated into the models. As always improved algorithms are needed to speed up calculation time. Uncertainties need a close analysis also.

Concerning the fuel kernel, thermodynamic data are scarce and oxygen potential measurements for a variety of actinide compounds are required, if further chemical buffering design of the kernel is to be achieved. The reduction of CO is an important parameter, thus its generation mechanism and its abatement are important issues to determine

## 1.4 Work Package 3

### 1.4.1 Introduction and reminder of objectives

PUMA has investigated the potential of High Temperature Reactor (HTR) and Very High Temperature Reactor (VHTR) technologies as a possible means of burning plutonium and minor actinides. HTRs are thermal reactors which are moderated by graphite and cooled by helium. As their name suggests, the chemical inertness of helium is used to advantage in HTRs to support coolant outlet temperatures in the region of 900°C, which allows very high thermal efficiencies to be attained in electrical generation. These temperatures are also very useful for process heat applications.

VHTRs would operate at temperatures in the region of 1000°C or more, which would allow the efficient generation of hydrogen for transport. These high temperatures necessitate the use of all-ceramic fuels to avoid the temperature limitations associated with the metallic alloy fuel cladding and structural materials used in conventional fuel assemblies.

While the current generation of nuclear power plants are dominated by Light Water Reactors (LWRs), (V)HTRs potentially offer superior performance in term of efficiency and economy, inherent safety and possibly in proliferation resistance. If these advantages can be realised in practice, (V)HTRs could be competitive with LWRs for the next generation of reactor build. If (V)HTRs successfully penetrate the power reactor market, it is important to understand how they could be used to manage historic arisings of separated plutonium and potential future arisings of minor actinides (MAs), and this is one of the main objectives of PUMA.

This section combines the main outcomes of the various investigations undertaken within Work Package 3, focused on the analysis of the (V)HTR's impact on the entire nuclear fuel cycle and economics. The main findings of the technical, (V)HTR's role in a future nuclear energy park, the implications on fuel cycle, economics and socio-political aspects are summarized in what follows.

The deliverables from WP3 are listed here. Their output is summarised in this report.

No.	Title	Responsible
D311	Report on technical characterisation of (V)HTR technologies	NNL (Hesketh)
D312	Report on economic characterisation of (V)HTR technologies	NNC (Millington)
D313	Report on environmental characterisation of (V)HTR technologies	NRG (Hart)
D314	Report on socio-political characterisation of (V)HTR technologies	CIRTEN (Cerullo)
D321	Report on future (V)HTR roles in European nuclear energy system scenarios	NRG (van Heek)
D322	Report on integrated nuclear energy system analysis	LISTO (Van Den Durpel)

D323	Report on economic viability of European nuclear energy system scenarios including (V)HTRs	NNC (Millington)
D324	Report on environmental impact of (V)HTR scenarios	NRG (Hart)
D325	Report on socio-political impact of (V)HTR scenarios	CIRTEN (Cerullo)
D326	Report on benchmark results of ORION, OSIRIS and DANESS codes	NNL (Hesketh)
D331	Report on uncertainty/sensitivity analysis of (V)HTR scenarios	NRG (Hart / Van Heek)

#### 1.4.2 Technical aspects of (V)HTRs (D311)

There are several common technological areas that underlie VHTR and HTR particulate fuels, high temperature primary pressure circuit, high temperature helium coolant and power conversion systems, ancillary systems and fuel cycle. Coated particle fuel consist of ceramic microspheres of oxide, carbide or oxy-carbide surrounded by layers of pyrolytic graphite and SiC to contain the fission products embedded in a graphite matrix.

World-wide, a total of seven HTRs have been built and operated. They are all characterised by the use of helium as the heat transfer medium, graphite as the moderating material and coated particle fuel technology. There has been a recent revival of both prismatic and pebble bed HTR designs with the General Atomics Modular High Temperature Gas-Cooled Reactor (MHTR); the General Atomics Gas Turbine Modular Helium Reactor (GT-MHR); the ANTARES prismatic HTR in France; the Pebble Bed Modular Helium Reactor (PBMR) project in South Africa and the HTR-PM in China. Of these systems, PBMR is at a much more mature stage of development than the others, with firm plans for construction of a prototype plant. These systems are all projected to be economically competitive with LWRs and both offer benefits relating to inherent safety and proliferation resistance.

The historical experience of designing, building and operating HTRs provides a very sound starting point for future development. Many aspects of (V)HTR technology have already been demonstrated historically and operating regimes of the historic HTRs in many cases coincide or overlap with those that will be required for (V)HTR. Compared with some other Generation IV systems for which there is no prior experience, the historic HTR experience could well be expected to shorten the time required for development and deployment of (V)HTRs by 10 years or more.

##### *Particle Fuel Technology*

Particle fuel technology is central to (V)HTRs. Nuclear fuel accumulates fission products and transuranics under irradiation. In conventional reactors, the fission products (some of which are volatile) and transuranics are ultimately retained by a metallic fuel clad. However, it is difficult to envisage any metallic cladding material being able to withstand the very high temperatures (900 to 1000°C coolant temperature) of a (V)HTR. At these temperatures, only ceramic materials are feasible and this is the main rationale for particle fuels, which comprise very small (<1 mm diameter) microspheres of fuel oxide, oxy-carbide or carbide, surrounded by ceramic multi-layers. There are three multi-layers in the standard arrangement, which is

known as the TRISO fuel particle. The multi-layers perform the same containment role as the metal cladding in conventional fuel. The small size of the fuel microsphere also largely eliminates the large centre to edge temperature differential of conventional fuel pellets, thereby reducing the maximum fuel centre temperature.

#### *(V)HTR Power Conversion Systems*

Helium has many very favourable properties as a heat transfer medium for (V)HTRs: as an atomic gas molecular dissociation is not an issue and helium is stable to very high temperatures; as an inert element helium does not chemically react with the reactor materials; there is no phase transition and therefore no concern over two phase flow (as is the case in water reactors); helium has good heat transfer properties and finally helium has a low neutron capture cross-section and does not become activated. These properties make helium a suitable medium for both removing heat from the core and for direct power conversion using the Brayton cycle, though it does also suffer the disadvantages of low density (and therefore low convective driving term in natural circulation) and high speed of sound (which leads to rapid depressurisation following a primary circuit break).

#### *(V)HTR Safety Approach*

The safety of a nuclear reactor depends on: ensuring that the reactor can be made sub-critical following an initiating event and subsequently kept in a subcritical condition; ensuring that the decay heat produced in the core following shutdown is removed reliably such that damage to the fuel and core does not occur.

In (V)HTR, as there is no phase change possible in the coolant, there is no plausible mechanism for the core to return to criticality following a reactor trip. This contrasts with the situation for LWRs where there is the potential for changes in coolant density and/or phase to increase post-trip reactivity, thereby leading to re-criticality. The lack of a credible re-criticality mechanism represents a significant inherent safety characteristic of (V)HTR.

Following a reactor trip, the decay heat from fission products and actinides provides a significant heat load that in conventional reactors must be removed reliably to avoid damage to the fuel and core and the possible release of fission products. Immediately after the reactor has tripped, the decay heat loading is of the order of 7% of the prior thermal output. For a (V)HTR module of 600 MW<sub>th</sub>, this equates to about 40 MW and is very significant. Although the decay heat output diminishes quite rapidly in the first few hours after the reactor trip, it still remains very significant for many days and weeks thereafter. In conventional reactors active systems are required to maintain peak fuel temperatures below the threshold for fuel and core damage. To achieve the required reliability, multiple redundancy is needed, increasing complexity. As these decay heat removal systems are safety critical, they need to be designed, constructed and maintained to very high standards, thereby representing a major component of overall generating cost.

In contrast, (V)HTR is designed to remove decay heat by the passive mechanisms of conduction and natural convection. This is possible because of the much higher temperatures that the all-ceramic particle fuel can tolerate without failure. This provides a larger temperature differential to drive conduction and convection than would be possible in a conventional reactor system, increasing the passive heat removal capacity. The elimination of active decay heat removal represents a very considerable simplification that reduces construction and operating costs. The safety case analyses are also greatly simplified, as it is possible to concentrate on demonstrating safe performance in the worst possible fault sequence. Fission

product releases following the worst credible accident should be negligible, with no requirement for off-site or site evacuation.

Passive safety can be considered one of the major advantages of (V)HTR. In conventional reactors it is necessary to analyse accident sequences using probabilistic risk assessment (PSA) techniques. Accident sequences are systematically analysed using failure probabilities assigned to each failure node. Multiplying all the failure probabilities provides an estimate of the overall probability of a particular sequence occurring. In conventional reactors, a Design Basis is defined which determines the cut-off point at which an accident sequence does not need to be considered. The Design Basis seeks to balance the radiological consequences of fault sequence against the probability of that sequence occurring, so that more severe faults are only tolerated at low frequencies. Normally, the cut-off point is taken to apply to fault sequences with a probability of  $<10^{-5}$  or  $<10^{-6}$  per year, though consideration is given to sequences lower than the cut-off to ensure that there are no cliff-edge effects just outside the Design Basis.

The D311-report on technical description of (V)HTR-technology has highlighted many aspects of (V)HTR technology where these systems show very promising potential. Compared with the LWRs with which it will be competing in the market, (V)HTR technology has potentially many advantages: safety; economics; transuranic inventory in spent fuel; proliferation resistance and others. Another benefit is the potential of (V)HTRs for burning transuranics, which could be used to establish low radio toxicity fuel cycles. If these advantages can be realised in practice, it will be realistic to envisage (V)HTRs making inroads in commercial power production and perhaps eventually rivalling LWRs in world capacity.

### 1.4.3 Potential role for (V)HTRs (D312)

The traditional role for nuclear energy has been large-scale electricity generation. Some efforts have been made to enter other markets with power reactors, but so far not successful enough for repetition and expansion. Examples are ship propulsion (US and Germany), district heating (Switzerland) and desalination (India). However, due to higher oil and gas prices, carbon emission constraints and altered perception of environmental impact of nuclear power, this might change in the future. The Generation IV programme identifies a number of future applications for nuclear power besides electricity generation (see Figure 11), which is also reflected in the Strategic Research Agenda of the EU's Sustainable Nuclear Energy Technology Platform (Figure 12). Then, this limited experience can be of use as a technical basis.

The High Temperature Reactor has been conceived in the 1960s with the purpose of generating high coolant temperatures for either increased electric conversion efficiency or industrial applications. Until now, the latter has not materialized yet; past HTR prototypes and demonstration plants have been generating electricity only. The Japanese HTTR is the first prototype HTR whose heat will be used to produce hydrogen. The Chinese HTR-10 is besides electricity generation, also used for heating the HTR-10 plant buildings during the cold season.

Only after the current and past prototype and demonstration plants were designed, interest in a new application of HTR emerged: that of 'waste burning' with 'waste' meaning here the transuranic actinides discharged with the spent fuel from HTR or other reactors.

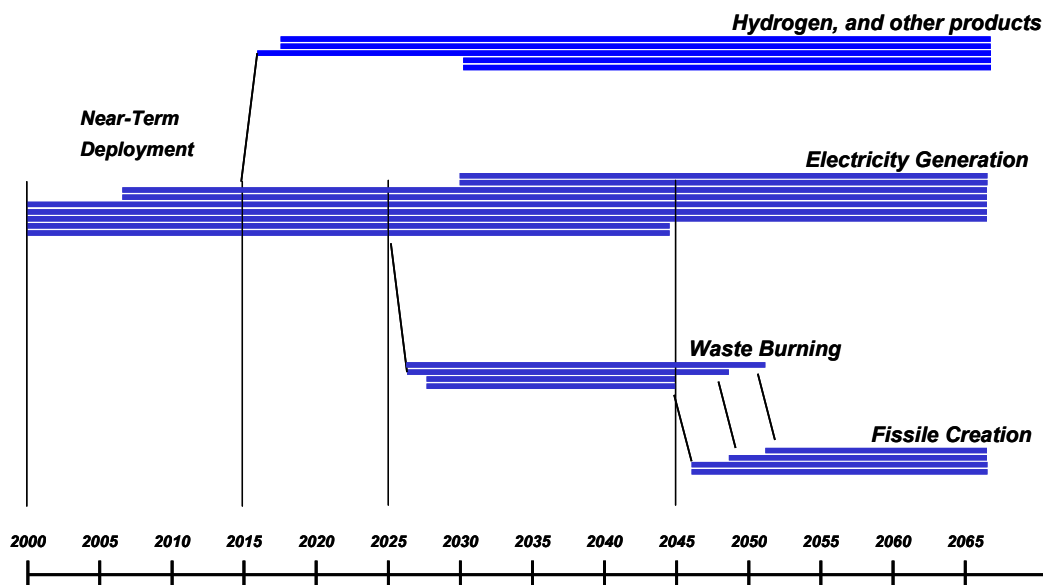


Figure 11: Nuclear energy applications according to the Generation IV Roadmap.

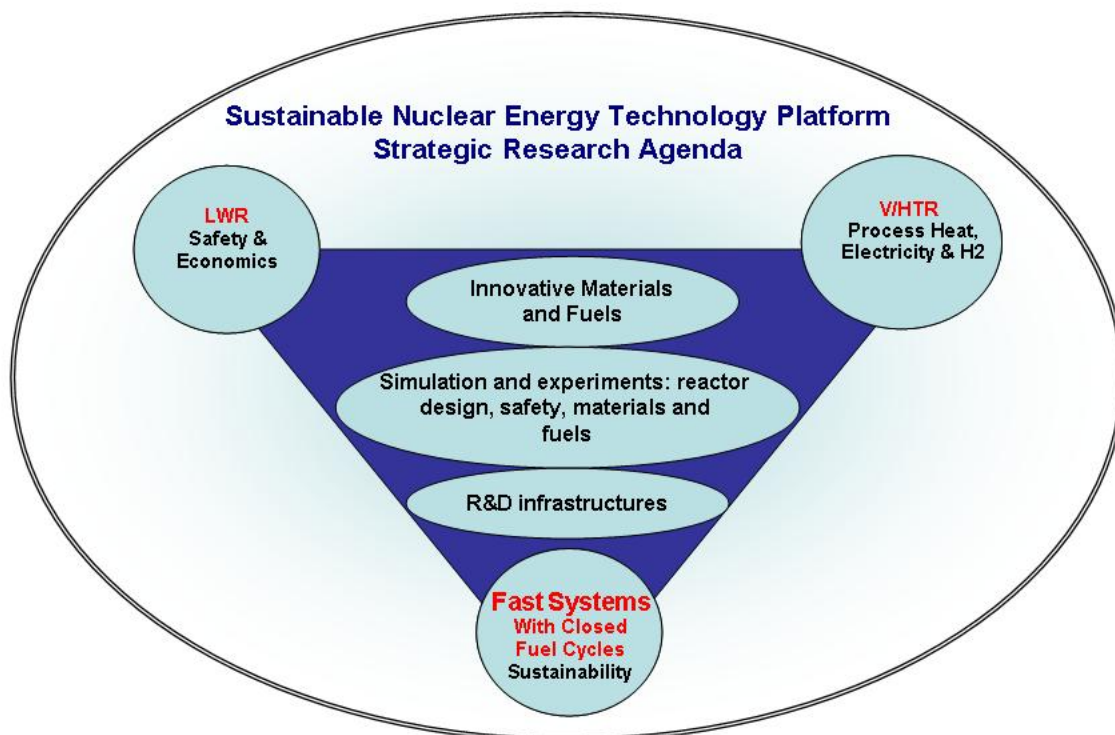


Figure 12: SNETP Strategic Research Agenda focus.

In report D312 all these applications are discussed briefly, followed by chapters on licensing aspects, future technical developments and market penetration.

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As a conclusion, the (Very) High Temperature Reactor is a reactor type with many potential applications. Whereas only electricity generation has been realized in practice with the early test and demonstration reactors, also non-electric energy products like process heat and hydrogen could become interesting in the future. This report gives an overview of the various processes for electricity generation and process heat production for selected products.

The fact that, in the past, electricity generation has been realized and non-electric energy production did not, cannot be attributed entirely to the low oil prices during the past few decades. Whereas electricity generation traditionally has been the business of electric utilities, companies with electricity generation as their core business, the situation with non-electric energy products is different. Until now, for companies in e.g. the (petro-) chemical sector, heat and electricity supply are relatively simple processes requiring limited investment, effort and manpower within the company. For nuclear energy supply, the investment will be significantly larger, as well as the complexity of licensing and operation. The trend of outsourcing of both electricity and heat supply in some industrial areas may be of help here.

In the report, it is assumed that this hurdle is overcome by 2020, and that introduction of non-electric nuclear energy products will start then. Two scenarios with a significant role for nuclear energy have been considered: one 'usual' growth scenario where demand will rise steeply during the whole 21<sup>st</sup> century mainly because of fossil phase-out and renewables not catching up, and one 'rise and fall' scenario, of nuclear capacity increasing in the first half of the century, but declining in the second half, being replaced by other technologies. These scenarios being used in the report D321, D324 and D331 as input to the systems analysis of (V)HTRs in EU27 nuclear energy future.

#### **1.4.4 Economic aspects of (V)HTRs (D312, D323)**

A series of dynamic simulations of mixed reactor scenarios have been performed, featuring LWRs and (V)HTRs. The aim of the study was to quantify and understand the cost penalties associated with the operation of HTRs in a TRU-burning mode. Based broadly on a simple linear nuclear capacity growth assumption within the EU27 states, four scenarios were selected as follows :

1. LWR once-through, followed by direct disposal
2. LWR & PBMR once-through, followed by direct disposal
3. LWR reprocessing and plutonium recycle in PUMA PBMRs
4. Plutonium / Minor Actinide recycle in PUMA PBMRs

For each of these scenarios, the levelised unit electricity cost has been derived for each reactor system and for the entire nuclear park. The price assumptions used to derive these cost estimates are based entirely on open literature sources and, in some cases, extrapolations and judgements. By their nature, all economic studies are very sensitive to assumptions and input data, particularly for advanced reactors for which little or no construction or operating experience is available, and for fuel cycle technologies which have



not been scaled up for industrial deployment. The conclusions reached in this study are liable to change as better cost data become available.

For a range of discount rates from 2% to 10%, the discounted generating cost levelised across the entire nuclear generating park demonstrates a slight cost penalty when plutonium burning in PUMA PBMRs is undertaken. This is around 0.05 Euro¢/kWh when compared with an LWR-only once-through scenario, and around 0.07 Euro¢/kWh compared with an LWR/PBMR once-through scenario. If long term back-end liabilities are assumed not to benefit from discounting, plutonium burning in PUMA PBMRs is the cheapest option, as back-end storage and disposal costs are reduced if Pu burning is undertaken. In each case, separated plutonium stocks are demonstrated to be minimised, resulting in clear proliferation and radio toxicity benefits. Burning of minor actinides along with plutonium results in a greater cost penalty, depending on discount rate, but this is based on more speculative fabrication price estimates.

As the main component of generating cost is due to repayment of the capital investment required for plant construction, which is typically around two-thirds of the total, the price estimates are most sensitive to assumed overnight capital costs.

In this respect, PBMR benefits from claims of capital construction costs which are significantly lower than the LWR. This assumption must be reconfirmed as the PBMR design matures and becomes available for deployment, and must be revised in any future studies of this nature.

Conventional fuel cycle costs typically account for ~15% of generating costs. They therefore have a smaller relative effect on total generating cost, unless costly new or additional processes are introduced. Fabrication of plutonium or plutonium/minor actinide coated particle fuel is such a process, and establishing robust industrial cost estimates is therefore identified as a key priority.

This study, together with the benchmark exercise conducted within PUMA WP3 has resulted in the establishment of detailed economic models which are available for future assessments as new cost data become available.

(V)HTRs have the potential to have lower overnight capital costs than LWRs due to the use of a direct power conversion cycle which removes the extra cost of the steam circuit. Additionally, the use of passive safety systems reduces the additional cost of some of the active safety systems associated with other plants. However, this cost gain is partially offset by the economies of duplication of key plant items when constructing modular reactors rather than a single large reactor, notably the reactor pressure vessel. The economics of modular construction does have advantages due to the phased construction programme which allows revenue to be generated earlier and therefore reduces IDC. The risk to the capital invested in a series of modular reactors is also lower, as single module unplanned outages will not affect other modules.

A comparison of fuel cycle costs reveals that the increased price of fabrication and higher uranium content of typical UO<sub>2</sub> HTR fuel is offset by the higher achievable burn-up, and the greater thermal efficiency of the direct power conversion cycle. The back-end fuel cycle costs are reduced relative to those of the LWR as the greater burn-up and thermal efficiency result in reduced spent fuel volumes per unit of generated electricity, and thus less interim storage and final repository capacity will be required.

In conclusion, the (V)HTR has the potential to compete commercially with the LWR, whilst performing the role of a plutonium and minor actinide burner. A quantitative assessment of a range of recycling scenarios will be performed in PUMA WP3, and the cost data presented in this report will form the basis of the economic aspects of the studies.

#### 1.4.5 Environmental aspects of (V)HTRs and associated fuel cycle (D313)

The environmental impact of (V)HTRs is mainly assessed using expert judgement on the expected life cycle inventory of such reactors and associated fuel cycle infrastructure needs. For that matter a literature review of life-cycle inventory studies has been undertaken to compose a Life-Cycle Inventory data-set to be used as a basis for further expert judgement in synergy with PUMA WP 1 and WP 2 activities.

The emphasis of the data is on the environmental impact of radionuclides that may be released during the different stages of the nuclear fuel cycle and that are potentially hazardous to the environment. These data have been obtained from a variety of sources which are mentioned in the respective sections. The life cycle inventory data were obtained both for LWRs, HTRs, and, as far as available, for sodium-cooled fast reactors (SFRs).

The releases of radioactive materials from the **conversion, enrichment, and fuel fabrication** plants (LWR fuel) are generally small and consist mainly of uranium series isotopes. For the model installations comprising the LWR fuel cycle, the normalized collective effective dose from these operations was estimated to be  $0.003 \text{ man Sv (GWa)}^{-1}$ . For the Th-fuel cycle this information could not be retrieved.

Radionuclides that are sufficiently long-lived and easily dispersed in the environment can give rise to global doses. The following radionuclides of specific interest in relation to the release from the different nuclear facilities:

##### Light Water Reactors

The radionuclides of specific interest in relation to the release from light-water reactors during normal operation are:

- H-3 (half life: 12.26 a), C-14 (5,730 a), Kr-85 (10.7 a) and I-129 ( $1.6 \cdot 10^7$  a).

##### Reprocessing Plants – LWR Fuel

The radionuclides of specific interest in relation to the release from LWR fuel reprocessing plants are:

- for the gaseous effluents: H-3 (half life: 12.26 a), C-14 (5,730 a), Kr-85 (10.7 a), I-129 ( $1.6 \cdot 10^7$  a), and Cs-137 (30.2 a);
- for the liquid effluents: H-3 (half life: 12.26 a), C-14 (5,730 a), Sr-90 (29.1 a), Ru-106 (1.02 a), and Cs-137 (30.2 a).

##### Gas-Cooled Reactors

The radionuclides of specific interest in relation to the release from gas-cooled reactors are:

- for the gaseous effluents: H-3 (half life: 12.26 a), C-14 (5,730 a), Ar-41, Kr-85 (10.7 a), and to a lesser extent Xe-133 (5.243 days);
- for the liquid effluents: H-3 (half life: 12.26 a);
- for the solid waste: H-3 (half life: 12.26 a), C-14 (5,730 a).

Based upon the assessment of all data it is apparent that C-14 makes an important contribution to the total radiological impact associated with nuclear power plants and reprocessing plants. It is therefore an important radionuclide when performing global optimisation studies of the nuclear fuel cycle and when comparing the radiological impact associated with different fuel cycles.

The impacts on both the local and global populations associated with the operation of an individual LWR or HTR, or the reprocessing of the fuel from a single reactor are smaller than the impact from natural C-14 and thus significantly smaller than the total impact from natural background radiation. With the possible exception of the maximum individual dose to individuals near reprocessing facilities this is also true for the combined facilities composing an LWR or HTR park equivalent to the current installed European nuclear capacity.

Assuming no retention of C-14, the C-14 releases from facilities in a once-through HTR cycle are significantly lower (by approximately one order of magnitude) than the releases from the facilities in a once through LWR cycle. For closed fuel cycles, incorporating reprocessing, the C-14 releases from facilities in the HTR cycle are higher (by approximately a factor 5) than those from facilities in the LWR cycle.

#### **1.4.6 Socio-political aspects of (V)HTRs (D314)**

(V)HTR, as one of the six Gen-IV facilities, has some specific benefits with respect to public perception, due to its original kind of fuel and to its innovative design features.

The analysis of Eurobarometer shows that the most important thing that enables the aversion to nuclear source are the waste (quantity, management, etc.). Another aspect is related to the safety operation of reactor, even if, as shows during the description of social indicators, nuclear chain has the minor number of fatalities per GW<sub>yr</sub> if compared with all the energy sources. In order to ensure a full public acceptance of the (V)HTRs, it is mandatory to analyze the non-proliferation aspects related to this kind of reactors. These issues are analyzed and discussed in Part II of D314.

For these reasons consecutively we describe only the advantages of (V)HTR with respect to safety and waste management, giving some additional notes also about economic point of view.

##### *(V)HTRs advantages concerning Safety*

If we look at the safety aspects of these reactors, we can note some peculiarities that makes the (V)HTR very attractive from "public" point of view. In fact, one of the more significant opposition of NPP is the release of fissions products under accident conditions. This opposition comes from the collective risks related to the accident occurrence.

Concerning the HTRs, heat-up experiments for accident simulation showed that fission products in Coated Particles (CPs) are completely retained up to rather high temperatures in the range of 2000°C. Retention of all the fission products in the fuel matrix, characterized by the presence of Coated Particles, is guaranteed in every accidental condition. In fact multi-layers perform the same containment role as the metal cladding in conventional fuel. The small size of the fuel microsphere also largely eliminates the large centre to edge temperature differential of conventional fuel pellets, thereby reducing the maximum fuel centre temperature.

The occurrence of air and/or water ingress into the core (conditions that would make not completely true the previous assumptions) are not possible in any case in the present design,

where the plant is laid underground). This capability finally lead to the development of (V)HTRs which are designed, even without any specific DHR system, in a way that the normal operating temperatures are in the region of 1250°C and even in worst case accident scenarios, the core designs are such that the fuel temperature will not exceed 1600°C. This is due to:

- a. the low power density (typically less than 10 kW/l)
- b. the high heat capacity of the great amount of graphite present into the core (slow transient responses);
- c. the high core height/diameter ratio (Radius = 1.85 m, Height = 11.0 m for PBMR);
- d. the possibility (even in the case of depressurization) of inherent passive decay heat removal capability by conduction to the soil (the reactor containment is laid underground).

In addition it could be noted that this positive safety behaviour means simpler licensing and reduced staffing needs.

An other important aspect related to the safety is the maintenance of safety condition under accident occurrence. This means a sub-critical condition where the decay heat produced in the core following shutdown is removed reliably such that damage to the fuel and core does not occur. Relative to this (V)HTR doesn't present the coolant phase change, so, there is no plausible mechanism for the core to return to criticality following a reactor trip. This means that the lack of a credible re-criticality mechanism represents a significant inherent safety characteristic of (V)HTR. In addition if reactor trip happens (V)HTR is designed to remove decay heat by the passive mechanisms of conduction and natural convection (it is possible because of the much higher temperatures that the all-ceramic particle fuel can tolerate without failure). For this reason the elimination of active decay heat removal represents a very considerable simplification that reduces construction and operating costs.

#### *(V)HTRs advantages concerning Economy*

Looking at the economics point of view, at least two items are to be highlighted:

- These plants are characterized by a relevant increase in efficiency (from 34 %, a typical value for the "classical" LWR, to ~50 % due to very high gas temperature, about 900 °C, entering in turbine);
- These reactors showed a dramatic system simplification. If we look at the plants, we can see how many auxiliary components are no more necessary. Fewer systems and fewer parts significantly reduce the complexities of reactor systems

#### *(V)HTRs advantages concerning Environment impact*

Finally, last but not the least, it is important to outline the low environmental impacts of (V)HTRs. In fact we can highlight that these reactors are characterized by:

- A high plant efficiency that implies a reduced thermal pollution;
- The high core adaptability to many type of fuel compositions lead to the capability of burning Pu and MA and also of using DU and/or Th ("waste" material) as fertile (in combination with Pu, U, etc.): this characteristic allows at the same time to reduce the long term radio toxicity of the final waste and a better exploitation of the nuclear fuel.
- The possibility of hydrogen production without any CO<sub>2</sub> emission. We stress that a CO<sub>2</sub>-free hydrogen production in significant quantities (possible with VHTR) is the only way to make hydrogen a real environmental acceptable alternative to the fossil fuels in the transport.

To conclude it is important to highlight that it is demonstrated that (V)HTR owns all the peculiarities that make this kind of reactors very suitable also from "political" point of view.

Particularly its unique intrinsic safety features could make this reactor "acceptable" by public opinion without any deep technical analysis that requires a specific knowledge in the field of nuclear engineering. In addition this reactor, meanwhile producing energy, addresses very well the target of (nuclear) waste minimization and resources preservation.

#### *Non-proliferation*

The Non-Proliferation issues on a world-wide basis are primarily addressed by the Treaty on the Non-Proliferation of Nuclear Weapons. The Non-Proliferation issues in the frame of EU are mainly covered by ESARDA activities.

Regarding (V)HTRs, first of all it is important to highlight that practically all the available literature on non-proliferation issues related to these reactors deals with pebble-bed reactors: this is mainly due to the fact that the continuous recycling fuel cycle (option not available for the block type HTR) makes this kind of HTR potentially more attractive for proliferation use.

In fact, among the (V)HTRs, the pebble-bed reactor (PBR) concept is often criticized as a potentially highly proliferating machine due to its online refuelling operation and because of the small size of its fuel elements (pebbles). The principal criticism is that online refuelling and its frequency (many pebbles every day), could make the inclusion of target pebbles possible. Such target pebbles would then be diverted to extract weapons-grade plutonium from them.

The scenario of greatest concern seems to be a dual use of the reactor by a country with no significant fuel cycle facilities of its own. In fact, a country with advanced fuel cycle facilities could produce the materials for a nuclear bomb via more effective and efficient means (e.g. using ad-hoc enrichment plants), so this scenario was not considered.

The following is a summary of the non-proliferation attributes of the PBR design:

- Low enriched fuel is used
- A closed system for fuelling and de-fuelling with on-line tracking of fuel or graphite sphere location. This reduces the possibility of clandestine introduction of target material or the protracted diversion of core nuclear material
- Only limited excess reactivity is allowed to overcome the temperature effects and to provide for equilibrium and transient fission product poisoning. The clandestine introduction of neutron absorbing target material (fertile) into the core will upset this reactivity balance and will have a noticeable effect on the core physics
- The PBMR is designed to store all the spent fuel generated during the operational lifetime of the reactor in the facility

- Should reprocessing become a viable option, the high burn-up achieved by the PBR fuel produces a mix of plutonium isotopes that does not allow the production of a reliable nuclear explosive device. In addition, the heat (and gamma rays) generated by these isotopes will cause rapid degradation of the high explosive components of such a device.

*A more in-depth analysis of proliferation resistance was part of Work Package 1 (D132).*

#### **1.4.7 Comparison of scenarios for impact analysis on nuclear fuel cycle (D321, D322, D324, D331)**

The introduction of (V)HTRs in EU27's nuclear energy system will demand not only R&D to develop the (V)HTRs including the appropriate fuels but also go associated with necessary changes in the nuclear fuel cycle allowing to fabricate the fuels for (V)HTRs and thus also assuring the availability of the fissile materials feeding such (V)HTRs all depending on their mission in EU27's nuclear energy system. The report D321 addresses the potential future roles of (V)HTRs in nuclear energy systems highlighting their particular role to extend the use of nuclear energy into non-electric applications via use of the higher-temperature process heat though also, due to rather unique characteristics of (V)HTRs reactor physics and particle fuels, their potential to fulfil a fuel cycle mission by burning transuranics (TRU) and thus aimed at contributing to radioactive waste management through reduction of the TRU-load in these wastes.

Both roles may go hand-in-hand as might be the case in EU27's future nuclear energy system though the future of (V)HTRs is obviously not defined by this combined role.

The combined report D322+D324+D331 analyses a set of EU27 nuclear energy system deployments without or with the introduction of (V)HTRs and with or without the second role of TRU-burning. As any study introducing scenario studies, a word of caution is very appropriate. Such scenario studies do not project nor prescribe what the future of EU27' nuclear energy system would become. Scenario studies only intend to illustrate possible future avenues of nuclear energy system deployment and what such deployment would entail from a holistic perspective, i.e. fissile material inventories, fuel cycle facility deployment needs, waste arising, economics, etcetera. As such, they serve an educational illustrative goal of providing some assessed cases allowing for the reader to grasp the main conclusions and allowing to interpret these conclusions in other scenario cases in the future.

This report summarises the nuclear energy scenarios analysis work performed within the PUMA-project by providing the results of seven nuclear energy system futures for EU27, ranging from a continuation of today's partial recycle in LWRs to a once-through option for LWRs and gradually towards the introduction of (V)HTRs and/or SFRs with these two reactor types fulfilling to varying degrees a TRU-burning role.

The report combines the reports D322, D324 and D331 into one report, given that the uncertainty/sensitivity-analysis (as initially to be reported in D331) bases itself upon the scenario-cases analysed in the D322-report addressing the major elements of the nuclear fuel cycle analysis relating to (V)HTR-introduction in EU's future nuclear energy system.

Figure 13 shows the comparison for the high energy demand scenarios of the total volume of disposed waste (SF and HLW) for the various nuclear energy system scenarios. The figure clearly shows the impact of reprocessing of the spent fuel and the corresponding reduction in volumes of long-lived and high-level radioactive waste resulting from this. The smaller though still important reduction of this waste volume between the Pu-management scenarios and the TRU-management scenarios needs to be put into perspective given the potential, though not

assessed due to shortage of detailed information, of additional medium-level waste arising in this scenarios as well as the higher amount of TRUs in Out-of-Pile as shown in Figure 14.

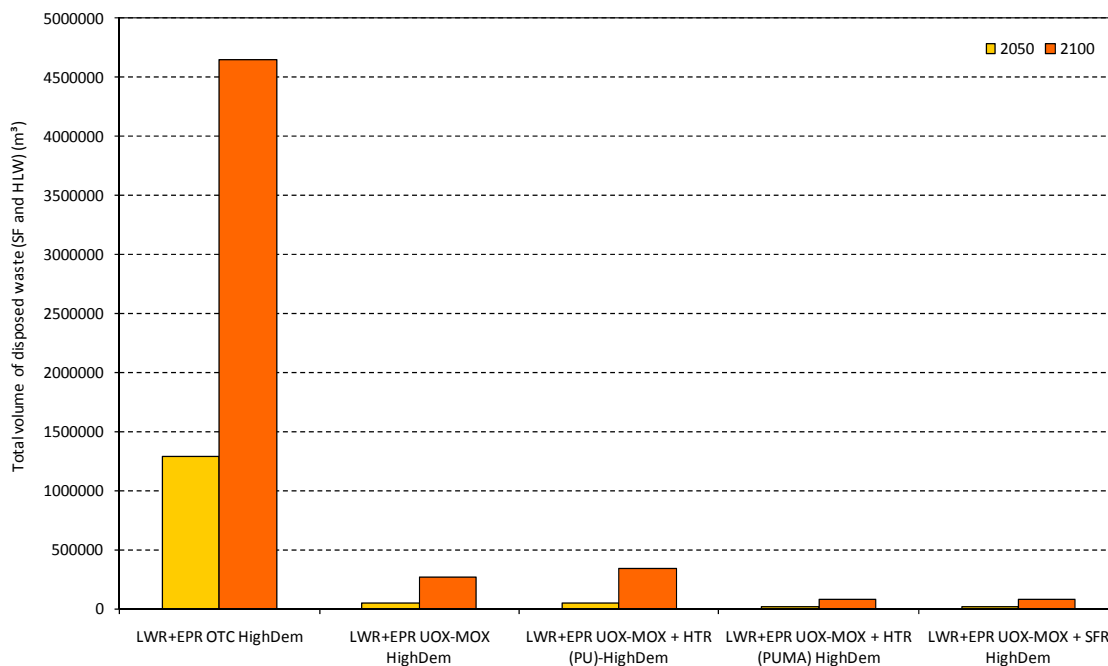


Figure 13: Comparison of disposed SF and HLW volumes for the different scenarios.

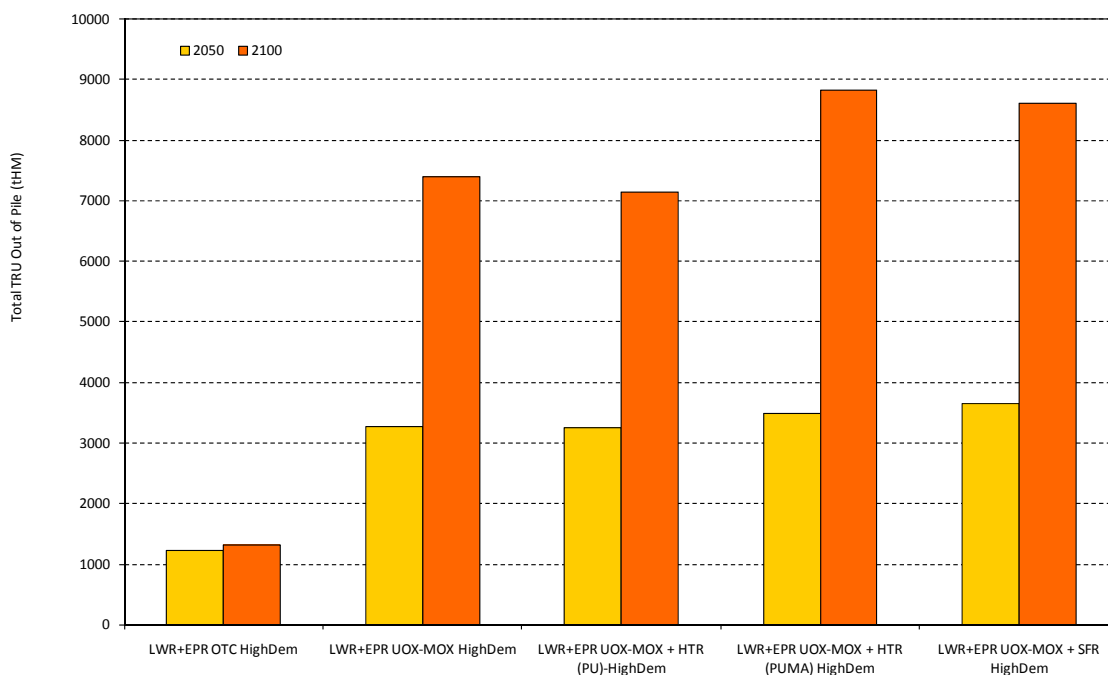


Figure 14: Comparison of Out-of-Pile TRU-amounts for the different scenarios.

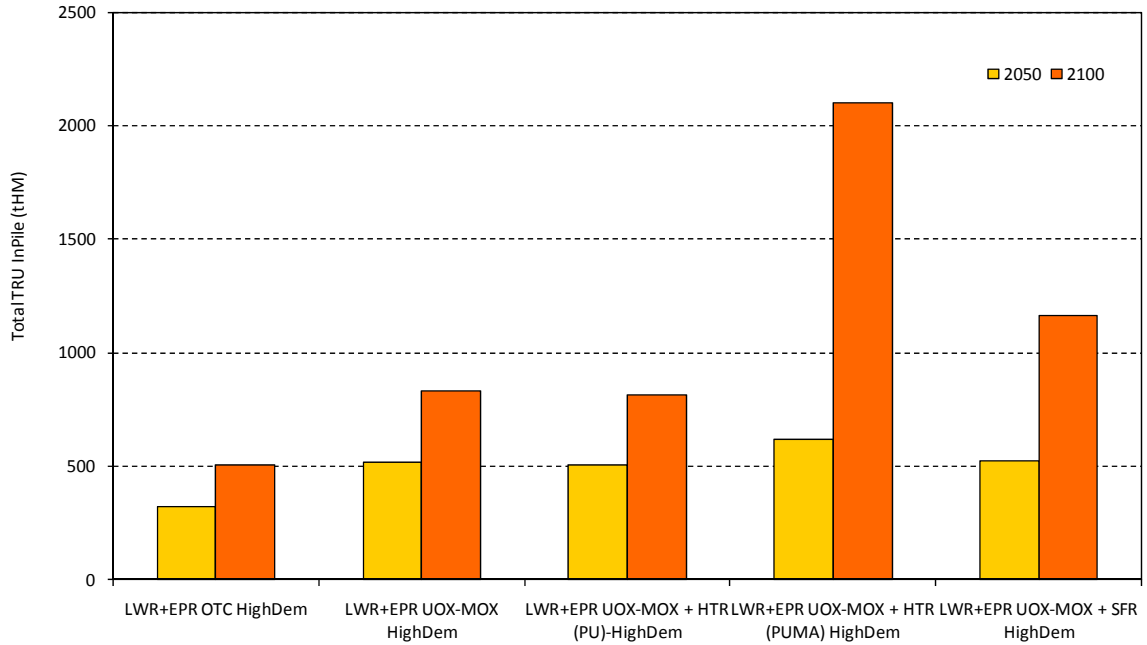


Figure 15: In-Pile TRU amounts for different scenarios.

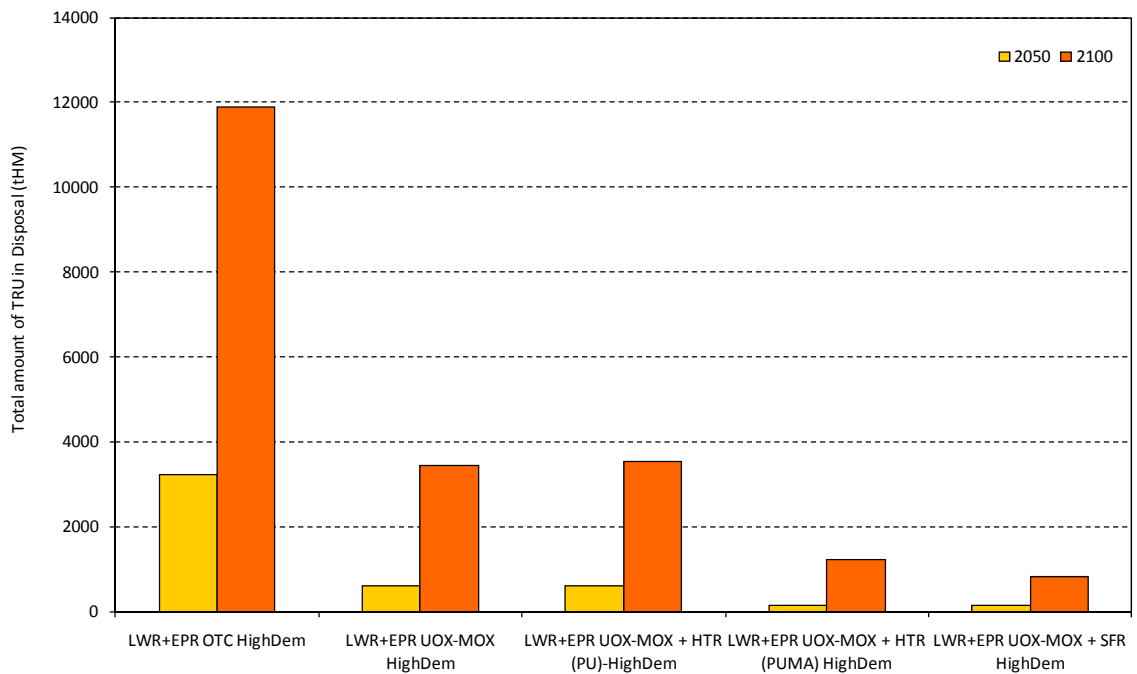


Figure 16: TRU-amount in disposed SF and HLW for different scenarios.



The higher burn-up and longer residence times of TRUs in (V)HTRs compared to FRs is shown in Figure 15 resulting into higher in-pile TRU-amounts for these scenarios with (V)HTRs. Though, the amount of TRU disposed in SF and HLW in repositories is smaller overall for FR-based scenarios compared to the (V)HTR-scenarios, as is shown in Figure 16.

Given that the park fraction for (V)HTRs and FRs in these scenarios remains overall limited, i.e. nuclear energy system essentially still governed by LWR/EPRs, the amount of  $U_{nat}$  savings is rather limited as shown in Figure 17. The impact on enrichment needs is however somewhat more visible as seen in Figure 18.

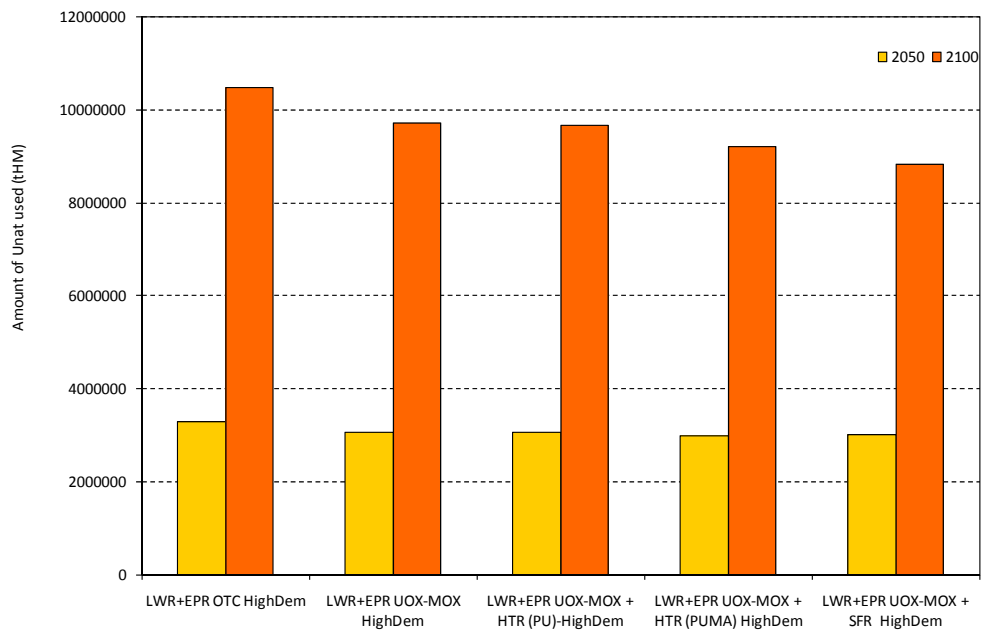


Figure 17:  $U_{nat}$ -needs for different scenarios.

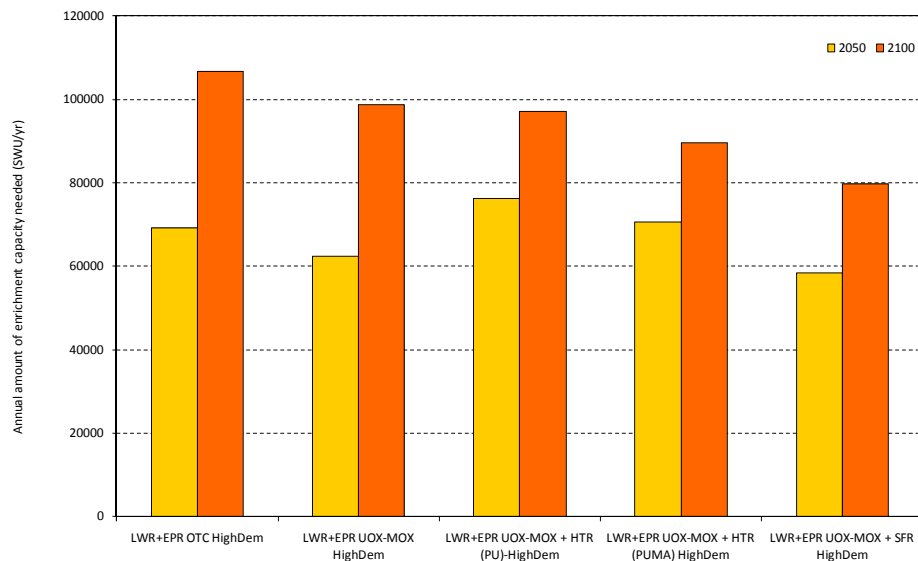


Figure 18: Enrichment needs for different scenarios.

Given the economic assumptions mentioned in Annex A of report D322+D324+D331, Figure 19 shows the average levelised energy generation costs (over the time period 2070-2100) for the scenarios where the comparison as such may only be made with the (V)HTR and FR-based scenarios as the costs for the existing LWRs in the first OTC-scenario were not explicitly accounted for and only new reactor plants were taken in the cost comparison. As can be seen from the figure, no specific cost difference given the assumed cost parameters is to be noted.

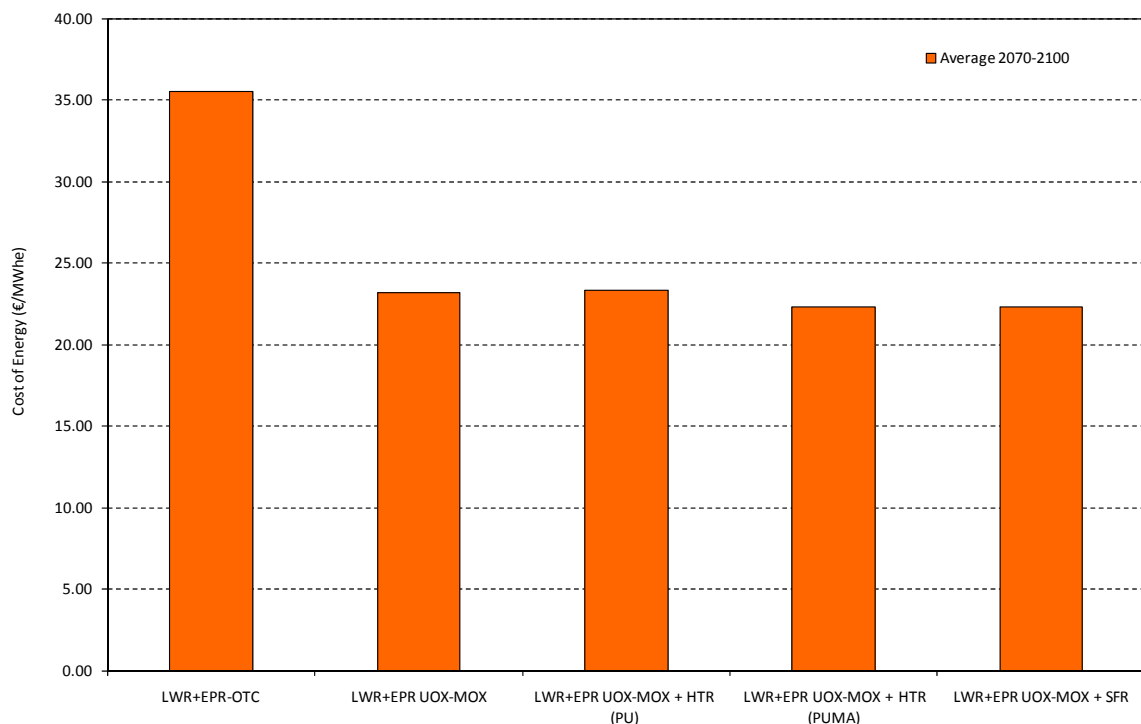


Figure 19: Averaged levelised energy generation costs for different scenarios.

#### 1.4.8 Benchmark of nuclear energy system codes (D326)

Reactor park simulations can be very complex and although they can in principle be modelled using spreadsheet applications, it is very difficult to do so. In a spreadsheet model it is not straightforward to incorporate all the constraints in a realistic fashion and it is difficult to achieve sufficiently flexibility to adapt the scenario. Furthermore, it is difficult to maintain adequate quality control of a complex spreadsheet representing a reactor park. As a result of such considerations, several independent reactor scenario modelling programs have been developed. Three such models are available to PUMA partners: ORION developed by Nexia Solutions; OSIRIS developed by AMEC and DANESS developed by LISTO. It is intended to use these programs to develop the European reactor park simulation and explore different deployment options for (V)HTRs.

This study has provided compared ORION, OSIRIS and DANESS models for the European Reactor Park scenario benchmark. The scenario is a very realistic one that has highlighted the

complexities that arise in real situations. This is thought to be the first occasion when scenario codes such as ORION, OSIRIS and DANESS have been compared for a realistic scenario.

The following points emerge from the comparison:

1. Although there are some differences between ORION and OSIRIS mass flows, for most nuclides the two codes agree reasonably closely. DANESS tends to agree less satisfactorily and generally shows a bias relative to the other two codes. While the various plants are still active, there are differences in the reported masses from the three codes, partly because the codes have nodalised the scenario slightly differently. In future comparisons, more effort should be devoted to ensuring the specification is sufficiently detailed to encompass the nodalisation, though in some cases it has to be accepted that the different codes may have limitations which prevent exactly equivalent nodalisations.
2. Some very large differences evident in the secondary parameters from ORION, OSIRIS and DANESS (i.e. decay heat output and radio toxicity). Exact agreement between the three codes could not reasonably be expected, given the differences seen in the basic material flows, but nevertheless the derived parameters are significantly more discrepant. Part of this is accounted for because the different codes track different nuclide sets. In ORION the nuclide set has been selected such that more than 99% of the decay heat output and radio toxicity contributions are captured and ORION is believed therefore to set a reliable reference point in this respect. The fact that OSIRIS and DANESS differ significantly suggest that these codes may not be as mature in this respect and may benefit from further development.
3. The benchmark has highlighted how important differences in the final nuclide inventories can arise from differences in the approaches used to interpolate for first cycle and last cycle fuel discharges. ORION and DANESS do not attempt any interpolation (i.e. for the last core it assumes all the fuel reaches the equilibrium discharge burn-up, which is not correct) and for some nuclides that saturate, this is the best approach. OSIRIS applies linear interpolation, which gives good results for nuclides which vary linearly with burn-up, but has results for saturating nuclides. In this scenario the differences can amount to a factor of 2 for some nuclides. This highlights the need for the implementation of more general interpolation methods for all three codes that more correctly account for the different nuclide behaviours.
4. An issue was highlighted that was not anticipated during the development of the benchmark specification. The specification defines the isotopic composition of LWR plutonium that is recycled in the HTRs. However, the decay of Pu-241 was not taken into account and indeed it is very difficult to do so because of the continually evolving situation in the scenario. Different approaches were adopted in ORION and DANESS on the one hand and OSIRIS on the other, with ORION and DANESS simply allowing the plutonium isotopics to evolve freely and OSIRIS freezing Pu-241 to force the plutonium isotopics to match the specification. The former approach is perhaps more realistic, but unavoidably deviates from the specification.
5. The scenario illustrates the potential value of HTRs in burning plutonium from the LWRs. The final plutonium mass is reduced by a significant amount, with the fissile plutonium being lower by a factor of about 3.5. This represents a benefit for proliferation resistance and in terms of the reduced radiotoxic inventory associated with plutonium.
6. The economic comparison shows close agreement between ORION and OSIRIS for the undiscounted case, but some discrepancies are found for the discounted case. Levelised costs are available for DANESS only for the undiscounted case, which agrees for the PWR component but not the HTR. Overall, the economic comparison was not satisfactory and suggests that further work is needed to ensure consistent and comparable approaches. This was the first application of the economics options in ORION and highlighted that further development is needed to make it workable.

Some of the conclusions above are not obvious and would never have emerged from a study with a single scenario code. This proves the value of the benchmark exercise and will allow the scenario codes to be applied with increased confidence in the future.

The study has illustrated that even for a well-defined scenario, it can be difficult to ensure the different scenario codes are modelling the scenario in precisely the same way and that more effort was needed than was available to fully resolve the remaining inconsistencies. The study has also highlighted unanticipated difficulties in the specification of the scenario itself, such as the treatment of Pu-241 decay and the treatment of non-equilibrium fuel discharges. With the inevitable differences in approach between the scenario codes, it is probably not possible to obtain complete agreement on every aspect of a scenario, but nevertheless the benchmark has proved to be very helpful in exposing the problem issues. If there is to be widespread deployment of HTRs, they will operate alongside the current LWR fleet and scenario codes such as ORION, OSIRIS and DANESS will be needed to provide a rigorous understanding of the interactions and synergies between the different reactor types.

## 1.5 Work Package 4

### 1.5.1 **Reminder of objectives**

Work Package 4 deals with project management and knowledge management. The first includes the usual management activities carried out to ensure proper delivery of the project according to objectives. The later includes both activities of communication / dissemination, and of education and training, with the objective of ensuring a maximum impact of the project's results.

The deliverables from WP4 are the following:

No.	Title	Responsible
D421	Project Quality Plan	LGI (Chauvet)
D431	Communication Action Plan	LGI (Chauvet)
D441	Plan of EUROCOURSE session in conjunction with RAPHAEL	USTUTT (Bernnat)

### 1.5.2 **Work performed, contractors involved and end results**

#### 1.5.2.1 Management and coordination

Project management and scientific coordination activities were essentially carried out by the coordinator (NRG), the project management office (LGI), and the WP Leaders (NRG, JRC, LISTO). All PUMA partners contributed when requested.

The management tasks were the following:

- **Contractual management:**
  - maintenance of the Contract and Consortium Agreement, in particular after the non-accession of one foreseen partner (Serco Assurance, UK), and the withdrawal of a partner after its cessation of activities (Belgonucleaire, Belgium)
  - periodic and final reporting to the European Commission
- **Financial management:** continuous funds monitoring, and minor budget reallocations during the course of the project
- **Quality management:** issue of the Project Quality Plan and monitoring of its proper application by all partners
- **Information management:** establishment of a web-based document management platform for sharing files and information between the partners; continuous management of the platform
- **Organisation of meetings:** seven plenary meetings took place over the period, systematically including Work Package (technical) meetings as well as Steering Committee and Executive Board (management) meetings; the list is given below.

**Meetings held during the project:**

Date	Location	Title	Minutes
16-17 Oct 2006	Petten	Kick-off meeting (WP, SC and EB meetings)	Issued
29-30 March 2007	Stuttgart	Plenary meetings n°2	Issued
19-20 Sept 2007	Brussels	Plenary meetings n°3	Issued
24-25 April 2008	Krakow	Plenary meetings n°4	Issued
8-9 Sept 2008	Karlsruhe	Plenary meetings n°5	Issued
16-17 March 2009	Knutsford	Plenary meetings n°6	Issued
9-10 July 2009	Amsterdam	Plenary meetings n°7	Issued
27-28 August 2009	Paris	Final Information Exchange Workshop (PUMA – US Deep Burn programme)	Issued

**1.5.2.2 Knowledge management, communication, and education & training**

Knowledge management comprises two types of activities:

- Communication and dissemination, essentially carried out by the coordinator (NRG), the project management office (LGI), the WP Leaders (NRG, JRC, LISTO), and several partners for specific actions.
- Education and training, organised by the University of Stuttgart (USTUTT)

This is detailed in the ***Final Plan for Using and Disseminating the Knowledge***.

## 2 Dissemination and use

Details of dissemination and communication actions are presented in the Final Plan for Using and Disseminating the Knowledge (FPUDK).

Given the nature of the PUMA results, and in view of the decision to provide them as European contribution to the international dialogue in the Generation IV International Forum (GIF), there are no publishable results apart from this report and those which have already been presented at conferences.