Pu and MA Management in Thermal HTR, QUO VADIS?

Insights from the Euratom PUMA project

J.C. Kuijper – NRG, Petten, The Netherlands

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Presented by F. Reitsma, IAEA

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AN ECN KEMA COMPANY
Coordinator
Leader WP1 / 4

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Science & Technology Consulting
Energy Business Consulting
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Generalities

- PUMA was a 3-year “STREP” from the EURATOM 6th Framework Programme (FP6)
  - Contract no. 036457, signed October 3, 2006
  - 1st September 2006 – 31st August 2009
- A consortium with **16** partners
  - From **9** countries (8 EU Member States + USA) + EC/JRC
  - Coordinated by **NRG** (The Netherlands)
- A budget of **3.580 M€** with EC funding of **1.850 M€**
  - **255** person-months of work
  - **50** deliverables

Technical objectives

Provide *additional* key elements for the use of contemporary and future (thermal) gas-cooled reactor designs as **burner for plutonium and minor actinides**. Focus on:

- Pu/MA HTR burner reactor physics & optimisation: **Reactor**
- Fuel design, manufacturing, performance modelling: **Fuel**
- Impact on fuel cycle and economics **Fuel cycle**

➔ Main question:

“Can the HTR be used as transmutation system for Pu and MA?”

PUMA addresses the improvement of sustainability of HTR/VHTR (reduce -production of- waste)

Work Packages

WP1

“Reactor”

“ Pu and MA transmutation/utilisation in HTR
& additional qualification of analysis tools ”

WP2

“Fuel”

“ Pu and MA fuel for HTRs ”

WP3

“Fuel cycle scenario”

“ Impact on economics and entire fuel cycle ”

WP4

“ Management, Communication
and Knowledge Transfer ”

PUMA

Interactions

- Exchange of information with other EU FP6 projects

RAPHAEL

GCFR
GAS COOLED FAST REACTOR

RED IMPACT

- Cooperation with international institutions

GEN IV International Forum

HTR-TN

AEN NEA

ISTC
МНТЦ

- Cooperation with US DOE project “**Deep Burn**”

WP1
Pu/MA transmutation/utilisation – full core simulation

NRG (lead), AGH, BN, EdF, IKE, KTH, UNIPI/CIRTEN, GA, NEXIA/NNC, FZJ, FANP

Objectives

- Demonstrate full potential of (contemporary) HTR designs to utilise/transmute Pu and MA fuel
- Identify necessary additional qualification of assessment tools (V&V)

Activities

- Definitions and analyses of reference (Pu-loaded) HTR systems and CP fuel:
  - **PBMR-400** with PuO_x fuel (cont. reload pebble bed)
  - **GT-MHR** with PuO_x fuel (prismatic block)
- Investigation of HTR core physics for Pu and MA fuel cycles, with emphasis on (transmutation) performance, within constraint of safe operation; optimisation of characteristics of fuel and reactor:
  - Pu/MA deep burn in pebble-bed HTR (incl. IMF)
  - Th/Pu fuel cycle in pebble-bed HTR
  - Pu/MA deep burn in prismatic HTR
  - Th/Pu fuel cycle in prismatic HTR
  - Reactivity transients of Pu/MA fuelled HTR (safety!)
  - Integrated LWR-HTR-GCFR symbiotic fuel cycles

WP1
Pu/MA transmutation/utilisation – full core simulation

Activities (cont.)

- Additional detailed investigations:
  - Calculation of helium/gaseous FP production in Pu/MA CPs
  - Assessment of proliferation resistance
- Miscellaneous:
  - Identify necessary (?) additional qualification of tools and opportunities to obtain experimental data to base it upon
  - Keep track of ISTC project #685.2 “HOT ASTRA”:

    Zero power critical facility at the Kurchatov Institute, Moscow. Pebble bed core (UOX fuel) with electrical heating to perform critical experiments at elevated temperatures. Possible future application: Pu-loaded pebbles.

Example: Study of Pu/MA-loaded pebble-bed HTR

Basic assumptions

- 400 MWth pebble bed HTR, similar to PBMR-400 (annular core)
- operating in a continuous reload (multi-pass) mode at a nominal power of 400 MWth;
- coolant inlet temperature: 773.15 K;
- coolant mass flow: 192.7 kg/s;
- coolant outlet temperature at nominal Hot Full Power conditions: 1173.15 K;
- outlet pressure at nominal Hot Full Power conditions: 9.0 Mpa

B.Y. Petrov, NRG

Also presented at PHYSOR 2012, Knoxville, TN,USA, 19 April 2012

Table I. Main System Dimensions

Description	Unit	Value
Equivalent core outer radius	m	1.85
Cylindrical height of the core (flattened core surface at the top and flat bottom reflector)	m	11.0
Radius of the central column graphite reflector	m	1.0
Effective height of the upper void cavity (from the levelled core surface to the bottom of top reflector)	m	0.5
Effective annular thickness of the side graphite reflector	m	0.9
Total height of top reflector	m	1.5
Total height of bottom reflector (distance from top of bottom plate to bottom of core)	m	4.0

- **number of fuel pebbles in the core: 452000**
- **total heavy metal loading per fuel pebble: 2.0 g**

Five fuel compositions have been considered:

- ***Pu*** **- plutonium;**
- ***PuMA*** **- plutonium and minor actinides;**
- ***Pu2xMA*** **- fuel composition with double minor actinide content compared to** ***PuMA*****;**
- ***IMX*** **- inert matrix fuel with isotope content identical to the one of** ***PuMA*****;**
- ***WP*** **- “wallpaper” fuel, in which the fuel is arranged in a spherical shell within the pebble, with the same isotope content as** ***PuMA*****.**

Number of coated particles per pebble:

- **fuel types** ***Pu*****,** ***PuMA*****,** ***Pu2xMA*****,** ***WP*****: 48816 coated particles**
- **fuel type** ***IMX*****: 26714 coated particles**

Table II. Coated Particle Dimensions and Initial Coated Particle Composition

Description	Unit	Value
Fuel kernel diameter – ***Pu***, ***PuMA***, ***Pu2xMA***, ***WP***	µm	200
Fuel kernel diameter – ***IMX***	µm	500
Thickness of kernel coating layers	µm	90 / 40 / 35 / 40
Kernel material type – ***Pu***, ***PuMA***, ***Pu2xMA***, ***WP***	-	$AnO_{1.7}$ *
Kernel material type – ***IMX***	-	$\left(\left(ZrO_2\right)_{0.84}\left(YO_{1.5}\right)_{0.16}\right)_{0.9}\left(AnO_2\right)_{0.1}$ *
Kernel coating material	-	C / C / SiC / C
Kernel material density – ***Pu***, ***PuMA***, ***Pu2xMA***, ***WP***	g/cm^3	10.89
Kernel material density – ***IMX***	g/cm^3	6.514
Density of kernel coating layers	g/cm^3	1.05 / 1.90 / 3.18 / 1.90

* *An* denotes “actinide”, i.e. Pu, Np, or Am (Cm is not present in the fresh fuel; it has been considered to be more efficient to separate Cm, let it decay to Pu and then recycle it into a reactor for further incineration.)

Table III. Fuel Pebble Characteristics

Description	Unit	Value
Fuel pebble outer radius	cm	3.0
Thickness of outer fuel-free zone	cm	0.5
Radius of central fuel-free zone – ***WP***	cm	2.2245
Total heavy metal loading per fuel pebble	g	2.0
Matrix density	g/cm^3	1.74
Packing fraction in pebble bed	%	61

Table IV. Fuel Composition

Nuclide	Fraction, wt % in ***Pu*** Fuel	Fraction, wt % in ***PuMA***, ***WP***, ***IMX*** Fuel	Fraction, wt % in ***Pu2xMA*** Fuel
Np-237	-	6.80	13.60
Pu-238	2.59	2.90	2.54
Pu-239	53.85	49.50	43.45
Pu-240	23.66	23.00	20.18
Pu-241	13.13	8.80	7.73
Pu-242	6.78	4.90	4.30
Am-241	-	2.80	5.60
Am-242m	-	0.02	0.04
Am-243	-	1.40	2.80

Computer codes used:

- **PANTHERMIX – the PANTHER code calculates the three-dimensional neutron flux and power distribution for a given temperature distribution, whereas the THERMIX-DIREKT code calculates the temperature and flow distribution for a given power distribution**
- **WIMS – generates a nuclear macroscopic cross section database for the PANTHER code**
- **FISPACT – calculates the nuclide inventory after a specified decay time following discharge of fuel.**

Fuel Used for One Full Power Year

Table V. Amount and Composition of Fuel Used in the System for One Full Power Year in Case of *Pu* Fuel

Isotope	Amount in Fresh Fuel, kg	Amount at 500 MWd/kg, kg	Amount at 600 MWd/kg, kg	Amount at 747.7 MWd/kg, kg	Amount 100 Years After Discharge, kg
Total Pu	1.953E+02	8.425E+01	6.260E+01	3.097E+01	3.446E+01
Total Actinides	1.953E+02	9.665E+01	7.696E+01	4.812E+01	4.793E+01

- **The total amount of plutonium isotopes in the discharged pebbles in the *Pu* case is 15.86% of their initial mass, while the total amount of actinides is reduced to 24.64%**
- **• The fissile fraction (i.e. the fraction of U-233, U-235, Pu-239, Pu-241, Am-242m, Cm-243, and Cm-245 in the total amount of actinides) decreases from 66.97% in the fresh fuel to 7.91% in the discharged fuel**

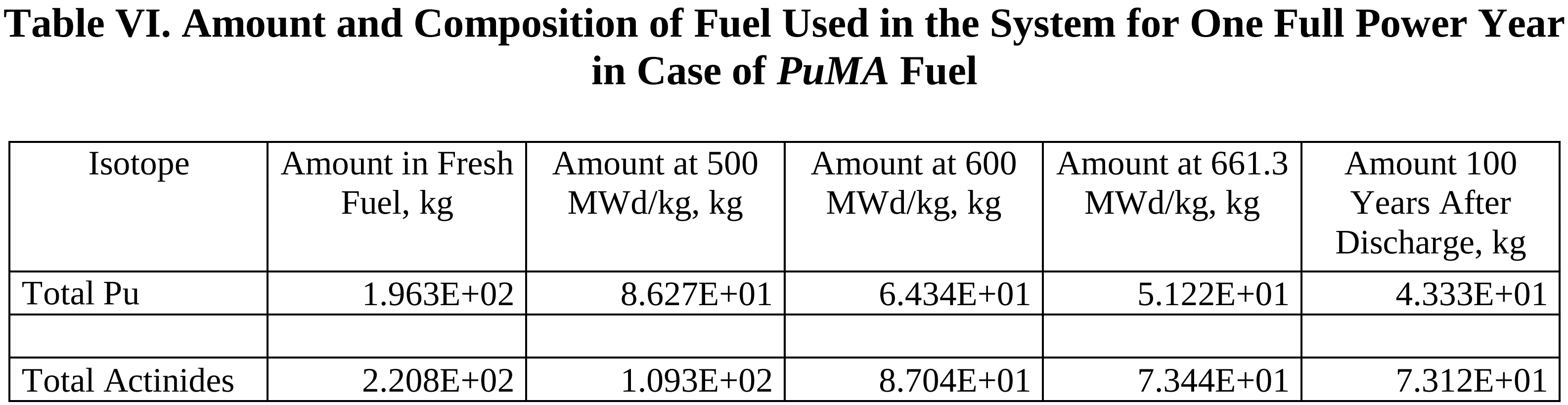


Table VI. Amount and Composition of Fuel Used in the System for One Full Power Year in Case of *PuMA* Fuel

Isotope	Amount in Fresh Fuel, kg	Amount at 500 MWd/kg, kg	Amount at 600 MWd/kg, kg	Amount at 661.3 MWd/kg, kg	Amount 100 Years After Discharge, kg
Total Pu	1.963E+02	8.627E+01	6.434E+01	5.122E+01	4.333E+01
Total Actinides	2.208E+02	1.093E+02	8.704E+01	7.344E+01	7.312E+01

- **The total amount of plutonium isotopes in the discharged *PuMA* pebbles is 26.09% of their initial mass and the total amount of actinides in the discharged fuel is 33.26% of the initial mass**
- **The fissile fraction decreases from 58.21% in the fresh fuel to 15.07% in the discharged fuel**

Table VII. Amount and Composition of Fuel Used in the System for One Full Power Year in Case of *Pu2xMA* Fuel

Isotope	Amount in Fresh Fuel, kg	Amount at 378.1 MWd/kg, kg	Amount 100 Years After Discharge, kg
Total Pu	3.007E+02	1.780E+02	1.166E+02
Total Actinides	3.861E+02	2.387E+02	2.380E+02

- **The total amount of plutonium isotopes in the discharged *Pu2xMA* pebbles is 59.17% of their initial mass and 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**

Table VIII. Amount and Composition of Fuel Used in the System for One Full Power Year in Case of *IMX* Fuel

Isotope	Amount in Fresh Fuel, kg	Amount at 500 MWd/kg, kg	Amount at 600 MWd/kg, kg	Amount at 692.6 MWd/kg, kg	Amount 100 Years After Discharge, kg
Total Pu	1.874E+02	8.105E+01	6.007E+01	4.135E+01	3.910E+01
Total Actinides	2.108E+02	1.043E+02	8.308E+01	6.346E+01	6.316E+01

- **The total amount of plutonium isotopes in the discharged *IMX* 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 from 58.21% in the fresh fuel to 10.10% in the discharged fuel**

Table IX. Amount and Composition of Fuel Used in the System for One Full Power Year in Case of *WP* Fuel

Isotope	Amount in Fresh Fuel, kg	Amount at 500 MWd/kg, kg	Amount at 600 MWd/kg, kg	Amount at 656.1 MWd/kg, kg	Amount 100 Years After Discharge, kg
Total Pu	1.979E+02	8.719E+01	6.507E+01	5.291E+01	4.415E+01
Total Actinides	2.225E+02	1.102E+02	8.773E+01	7.518E+01	7.486E+01

- **The total amount of plutonium isotopes in the discharged *WP* pebbles is reduced to 26.74% of their initial mass, while the total amount of actinides is reduced to 33.79%**
- **The fissile fraction decreases from 58.21% in the fresh fuel to 15.88% in the discharged fuel**

- After 100 years of decay following the fuel discharge, the total amount of actinides remains almost unchanged for all of the fuel types
- Among the Pu isotopes, only the amount of Pu-241 is reduced significantly due to its half-life of 14.35 y; this change is more pronounced in absolute numbers in cases of fuel compositions containing MA, as their content of Pu-241 at discharge is higher than the one for *Pu* fuel. This leads to a higher reduction of the total amount of Pu over the period of 100 years for fuel types which contain MA
- The quantity of Pu-238 decreases (half-life of 87.7 y), while the one of Pu-239 increases slightly (α-decay of Am-243 to Np-239 with half-life of 7370 y and β-decay of Np-239 to Pu-239 with half-life of 2.3565 d)
- The amount of Pu-240 increases due to α-decay of Cm-244 with half-life of 18.10 y. The quantity of Pu-242 practically remains the same due to its long half-life (373300 y)

Attained Discharge Burnup of the Fuel

Table X. Attained Discharge Burnup of the Fuel: Minimum Value

Fuel type	Unit	Value
Pu	MWd/kg	737.0
PuMA	MWd/kg	640.0
Pu2xMA	MWd/kg	347.0
IMX	MWd/kg	678.0
WP	MWd/kg	634.0

Table XI. Attained Discharge Burnup of the Fuel: Average Value

Fuel type	Unit	Value
Pu	MWd/kg	747.7
PuMA	MWd/kg	661.3
Pu2xMA	MWd/kg	378.1
IMX	MWd/kg	692.6
WP	MWd/kg	656.1

Local Volume-averaged Burn-up Distribution at Equilibrium

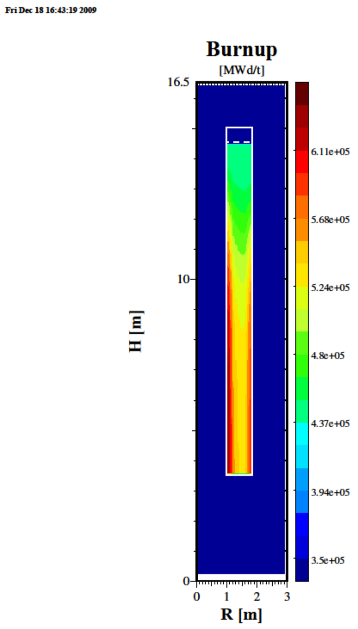


Figure 1. Volume-Averaged Burnup in Case of *Pu* Fuel

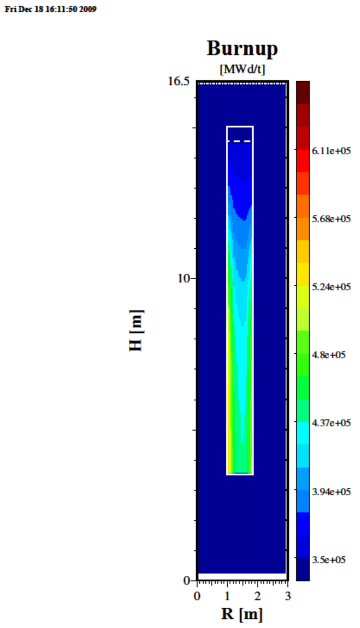


Figure 2. Volume-Averaged Burnup in Case of *PuMA* Fuel

In both cases, the maximum in the burnup distribution in radial direction is at the boundary of the inner reflector and the minimum is in the middle of the core

Power Density in the Core

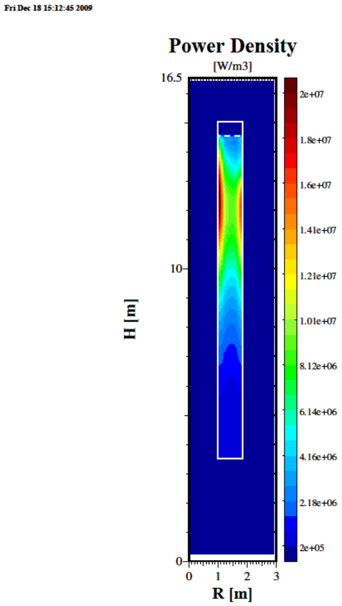


Figure 3. Power Density in Case of
Pu Fuel

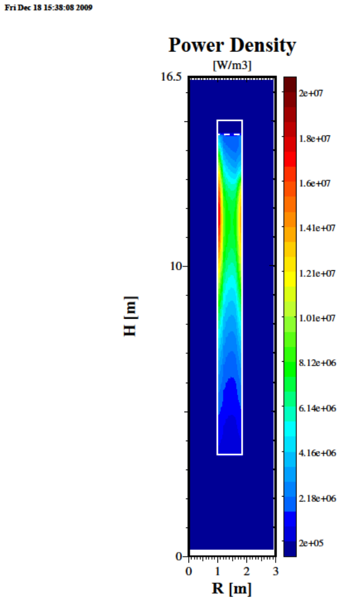


Figure 4. Power Density in Case of
PuMA Fuel

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

Power Density in the Core

The reactor fuelled by *Pu* features higher maximum values and larger axial gradient of power density than those fuelled by plutonium and minor actinides

Table XII. Power Density in the Core: Maximum Value

Fuel type	Unit	Value
Pu	W/m^3	1.996E+07
PuMA	W/m^3	1.718E+07
Pu2xMA	W/m^3	1.530E+07
IMX	W/m^3	1.749E+07
WP	W/m^3	1.707E+07

Pebble Surface Temperature

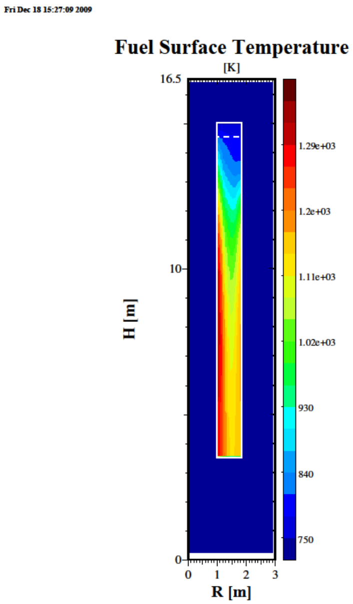


Figure 5. Fuel Surface Temperature
in Case of *Pu* Fuel

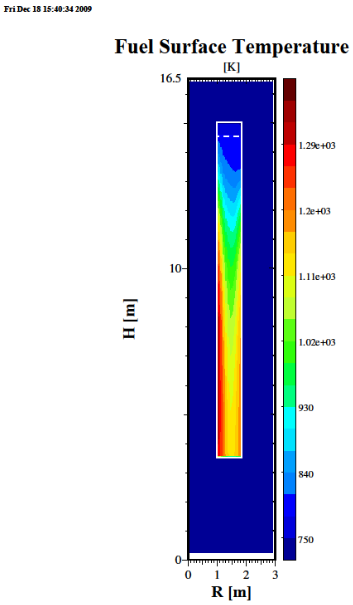


Figure 6. Fuel Surface Temperature
in Case of *PuMA* Fuel

For both types of fuel, the maximum pebble surface temperature is observed at the boundary of the inner reflector

Pebble Surface Temperature

Table XIII. Pebble Surface Temperature: Maximum Value

Fuel type	Unit	Value
Pu	K	1308.6
PuMA	K	1304.6
Pu2xMA	K	1346.6
IMX	K	1299.3
WP	K	1305.2

Temperature Reactivity Coefficients at Cold Zero Power

The reference state for a calculation at Cold Zero Power (CZP) is an isothermal reactor at 300 K, all control rods out, and zero xenon conditions

Table XIV. Temperature Reactivity Coefficients at Cold Zero Power

Fuel type	Unit	Value for Fuel and Moderator	Value for Reflectors
Pu	pcm/K	0.4610	4.0496
PuMA	pcm/K	1.0634	4.0008
Pu2xMA	pcm/K	-2.4535	4.7209
IMX	pcm/K	1.4615	3.9409
WP	pcm/K	1.1809	3.9940

The total temperature reactivity coefficients at CZP are positive in all of the cases; this fact means that reactor design changes are needed in order to improve feedback at low temperatures

Temperature Reactivity Coefficients at Hot Zero Power

The reference state in a Hot Zero Power (HZP) case is an isothermal reactor at nominal coolant inlet temperature (773.15 K), all control rods out, and zero xenon conditions

Table XV. Temperature Reactivity Coefficients at Hot Zero Power

Fuel type	Unit	Value for Fuel and Moderator	Value for Reflectors
Pu	pcm/K	-2.8106	2.3269
PuMA	pcm/K	-2.4840	2.2789
Pu2xMA	pcm/K	-4.1854	2.2052
IMX	pcm/K	-1.6839	2.2347
WP	pcm/K	-2.4159	2.2831

The total temperature reactivity coefficients at HZP 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.

Conclusions

- The results of this study 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

Additional remarks

- Further studies (e.g. within the “Deep Burn” project) indicate that a negative temperature reactivity coefficient can be obtained by using burnable poison.
- A well-tuned mixture of erbium and born burnable poison will also retain an acceptable discharge burn-up of the Pu or Pu/MA fuel
- The 400 MWth pebble-bed design is not the best choice for a Pu- or Pu/MA-utilising reactor, as the fuel temperature during normal operation is too high (>> 1250 degrees C). It is recommended to lower the reactor power.
- Except for water ingress no significant difference in transient behaviour was found compared with UO_2 cores, also not for DLOCA and PLOCA accidents.
- Similar findings arise from studies of the prismatic HTR

WP2
Pu and MA fuels for HTRs

JRC-ITU (lead), NRG, BN, General Atomics, JRC-ITU, KTH, NEXIA, TUD, FZJ

Objectives/Activities

- **Further development of fuel performance models appropriate to HTR CP fuel (incl. extension to Pu/MA fuel); development of helium and swelling behaviour models for coated Pu and MA particle fuels**
- **Establishment of helium behaviour in both Pu and MA kernels and PyC and SiC coating layers**
- **Characterisation of existing PuO_2 coated particles from BN**
- **Design of coated particles suitable for ultra high burn-up of Pu and MA kernels as impregnated Inert Matrix Fuel (IMF)**
- **Design of an irradiation facility for Pu/MA coated particles, meeting the requirements for a future irradiation test in the HFR Petten, NL**

WP3
Impact on economics and entire fuel cycle

LISTO (lead), NRG, NEXIA, FANP, KTH, GA, NNC, FZJ, CIRTEN

Objectives

- **Assess potential future roles for HTRs/VHTRs in delivering energy products, while performing a TRansUranium (TRU) management function in a nuclear reactor park**

Activities

- **Characterisation of HTR/VHTR technology, associated fuel cycle infrastructure and waste management technologies**
- **Assess the role(s) of HTRs/VHTRs with TRU management function; aspects:**
  - ***Technological impact***
    - **Technological feasibility**
    - **Focus on European nuclear reactor park**
  - ***Economic impact***
    - **Potential market penetration**
  - ***Environmental impact***
    - **Life cycle inventory analysis for HTRs**
    - **(Secondary) waste arising**
    - **Waste management options**
    - **Separated fissile material inventories**
    - **Losses in the nuclear fuel cycle, as well as into non-nuclear material streams;**
  - ***Socio-political impact***
    - **Proliferation risk**

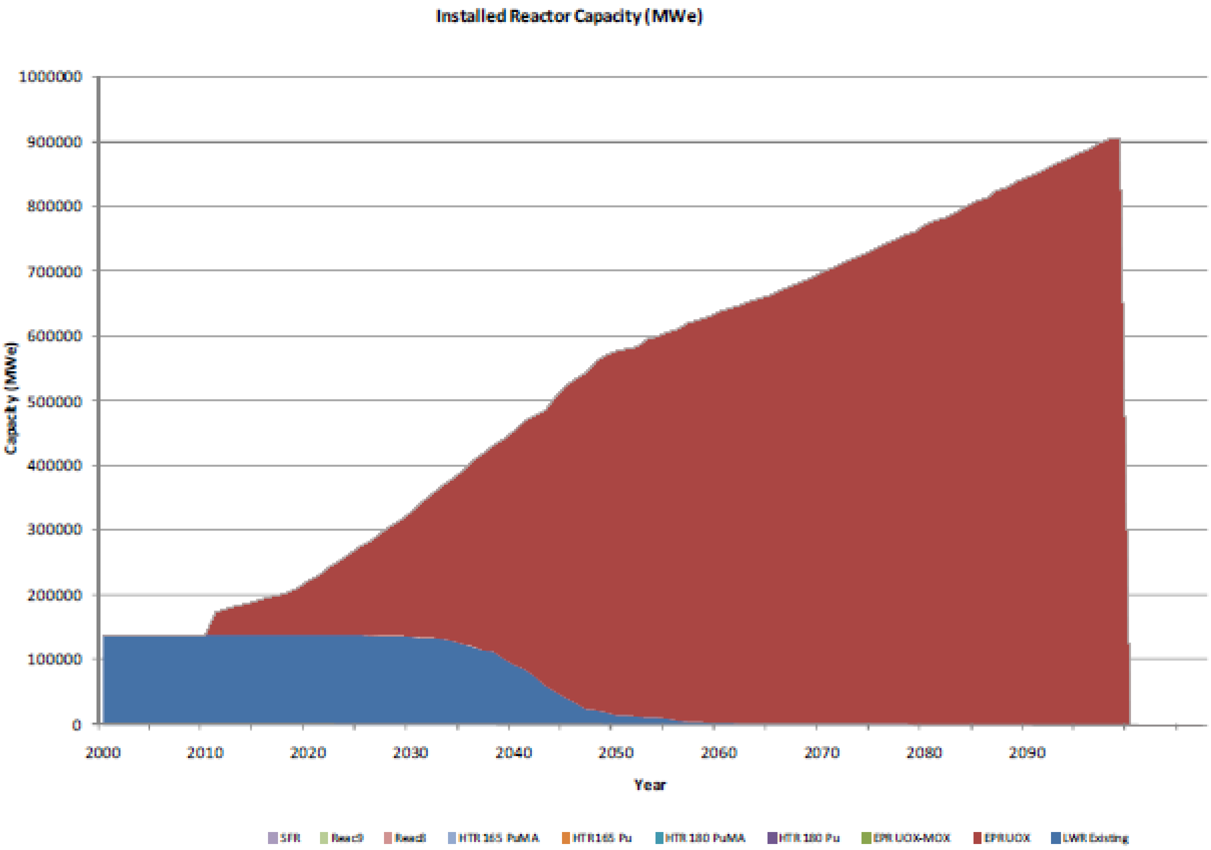
WP3
Impact on economics and entire fuel cycle

- Investigation/intercomparison of several scenarios, including sensitivity/uncertainty analysis:
  - European LWR park without HTR (reference)
  - European LWR park without HTR + Gen-III LWR with OTC and mono-Pu recycling
  - Mixed LWR and HTR park, HTR in OTC mode
  - Mixed LWR and HTR park, HTR as TRU burner
  - Mixed LWR, HTR and (S)FR reactor park
- Benchmarking of scenario analysis codes OSIRIS, ORION, DANESS

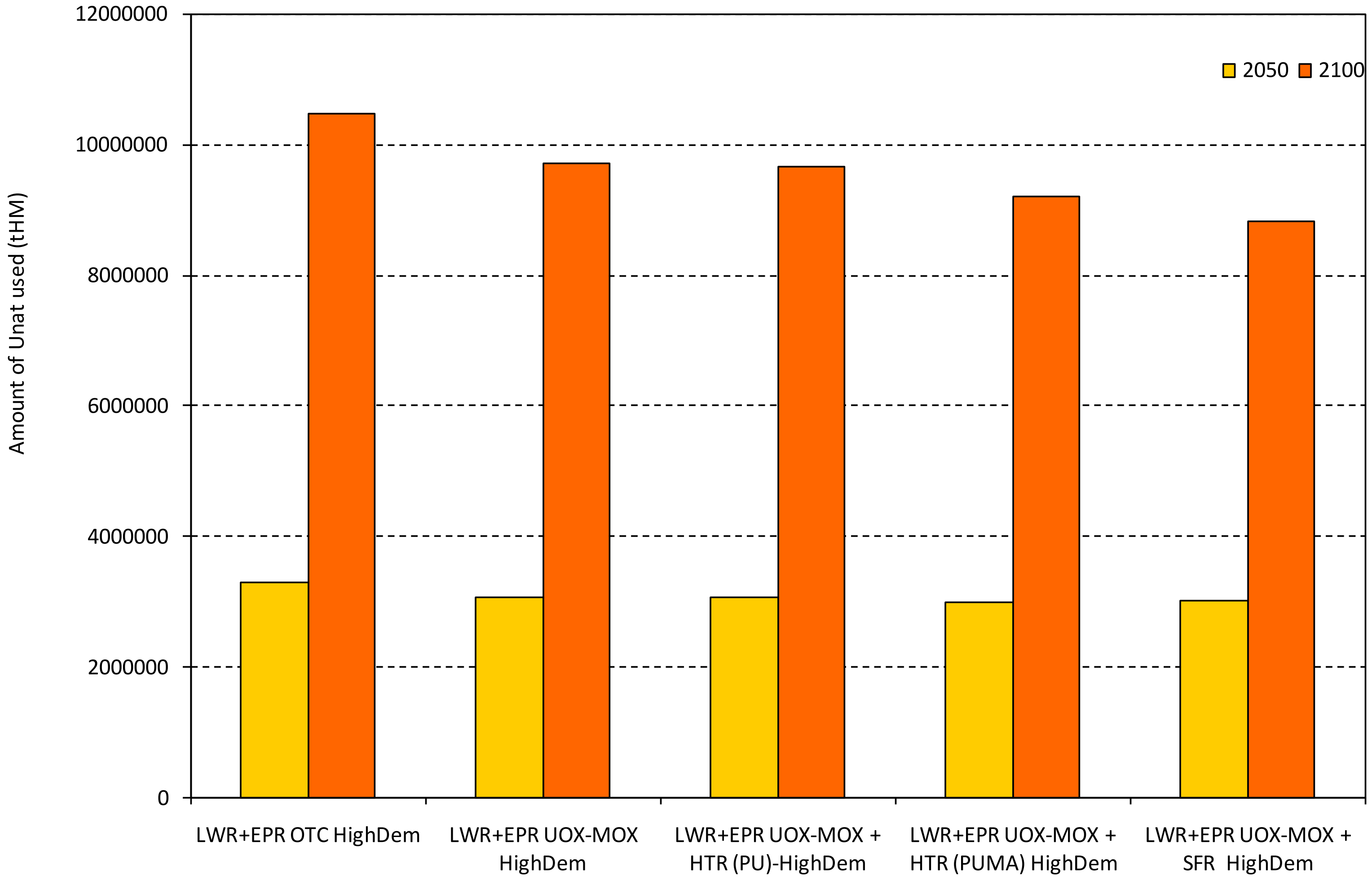
Example:
Impact of HTR on EU 27 fuel cycle

- Simulation with DANESS code
- Input: nuclear energy demand scenario (high demand variant)
- Initial situation: reality “today”: existing reactors, fuel fabrication facilities, reprocessing facilities, etc. in EU
- Simulation will deploy new facilities if necessary and possible within technical and economical constraints
- Large set of assumptions (see report…)
- Final situation: year 2100. Snapshots at 2050 en 2100
- Comparison of 5 scenarios (see previous slide)
- PuMA HTR properties from PUMA WP1

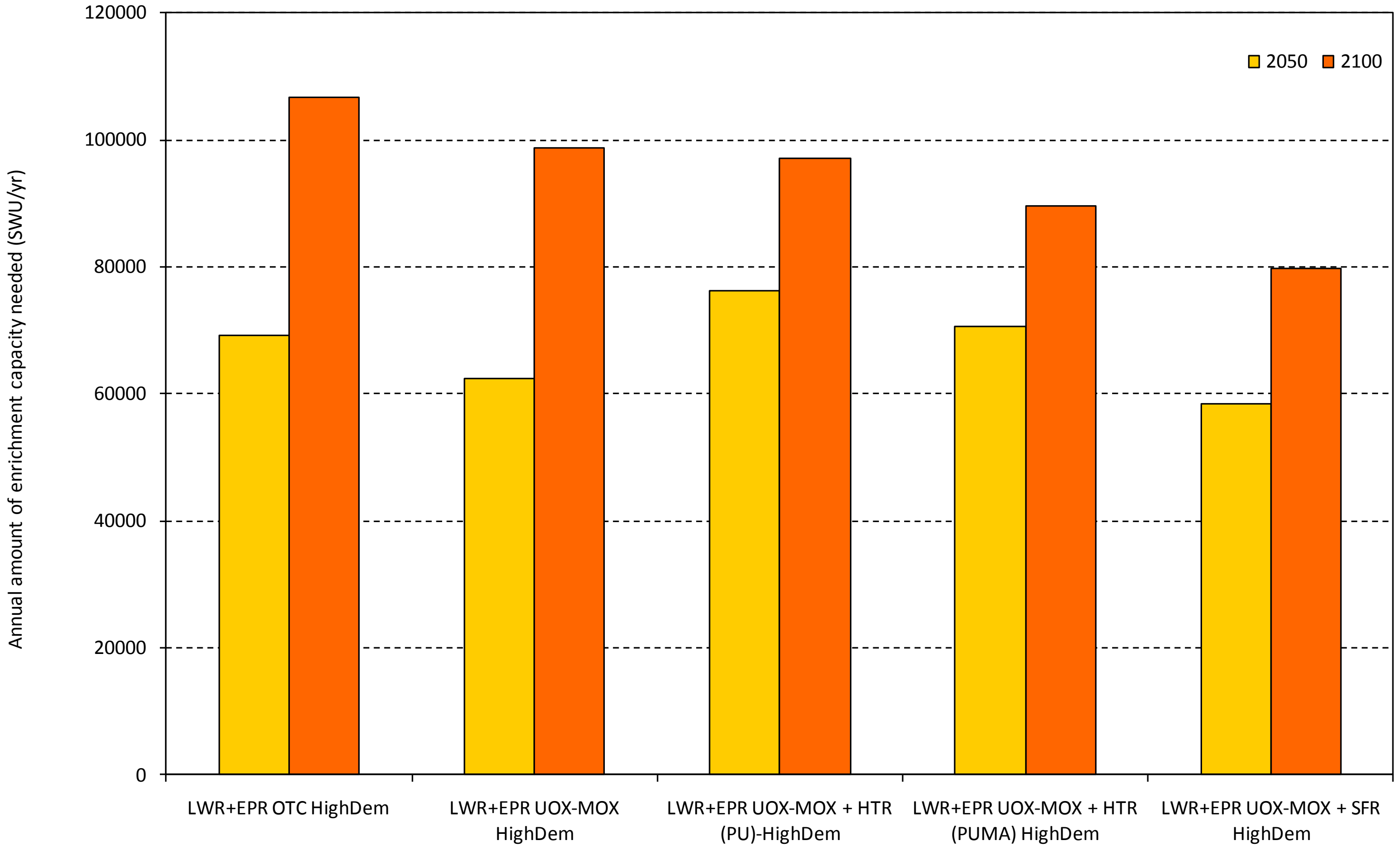
Input: Nuclear energy demand in EU 27 (high variant)



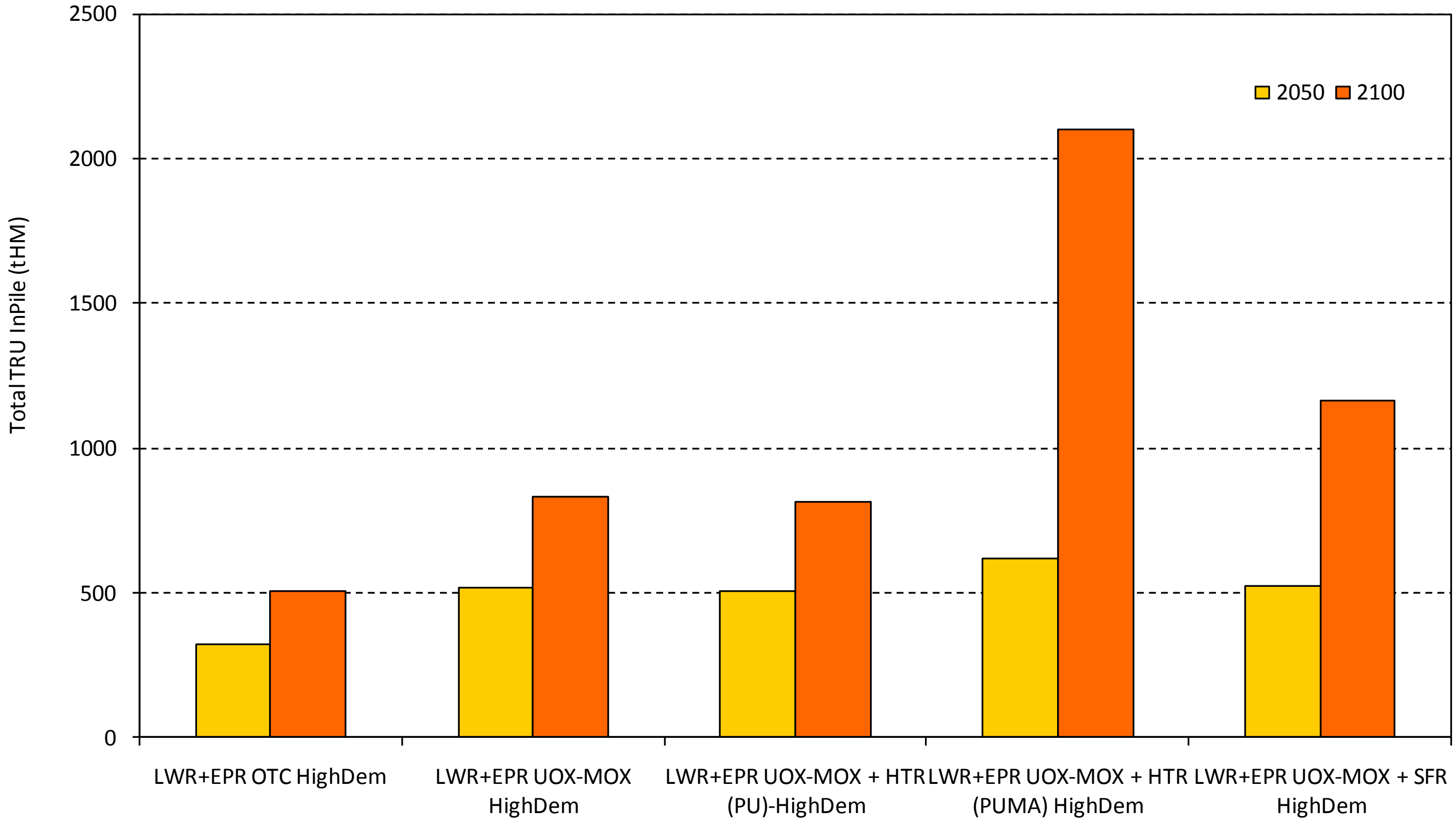
U_{nat}-needs for different scenarios.



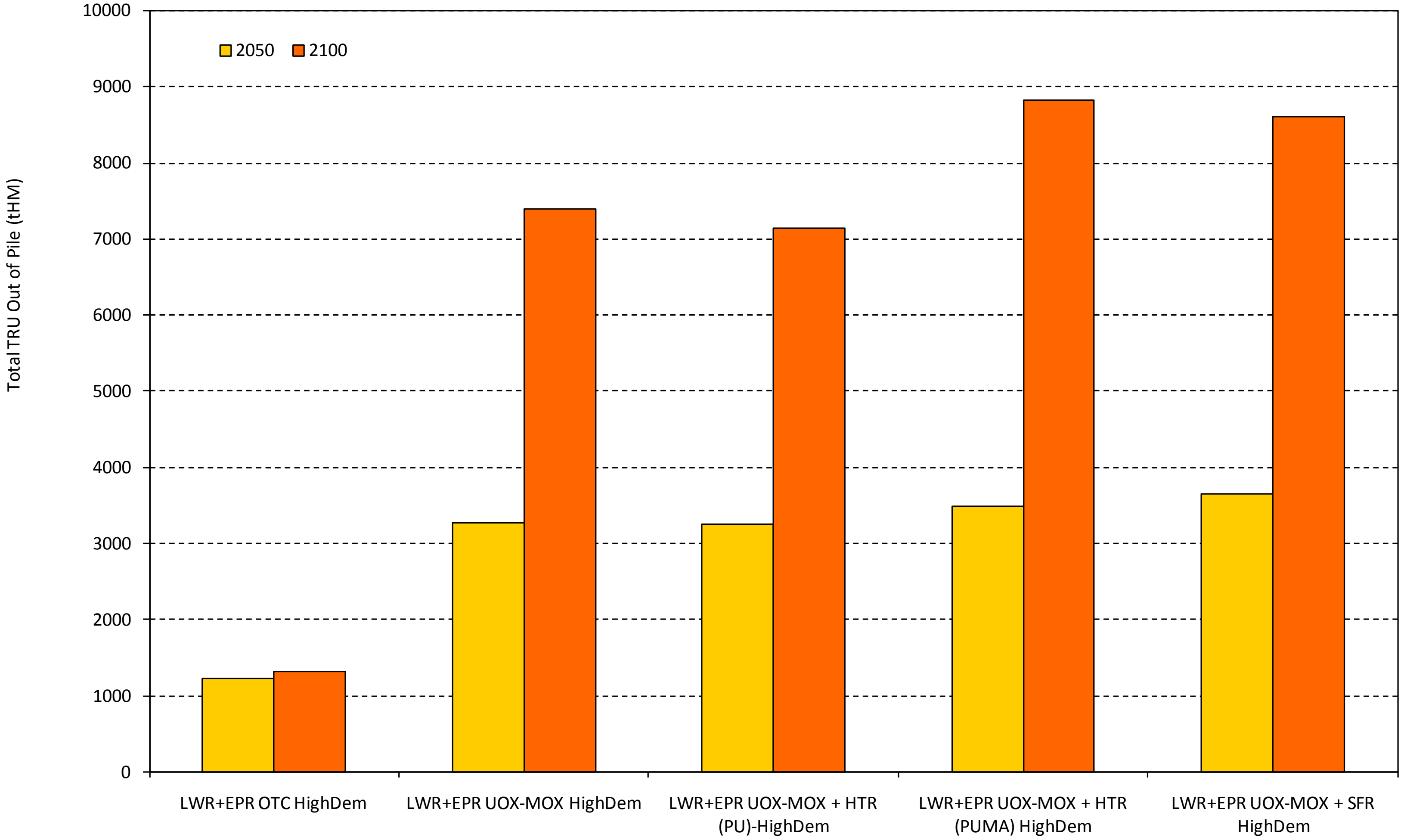
Enrichment needs for different scenarios



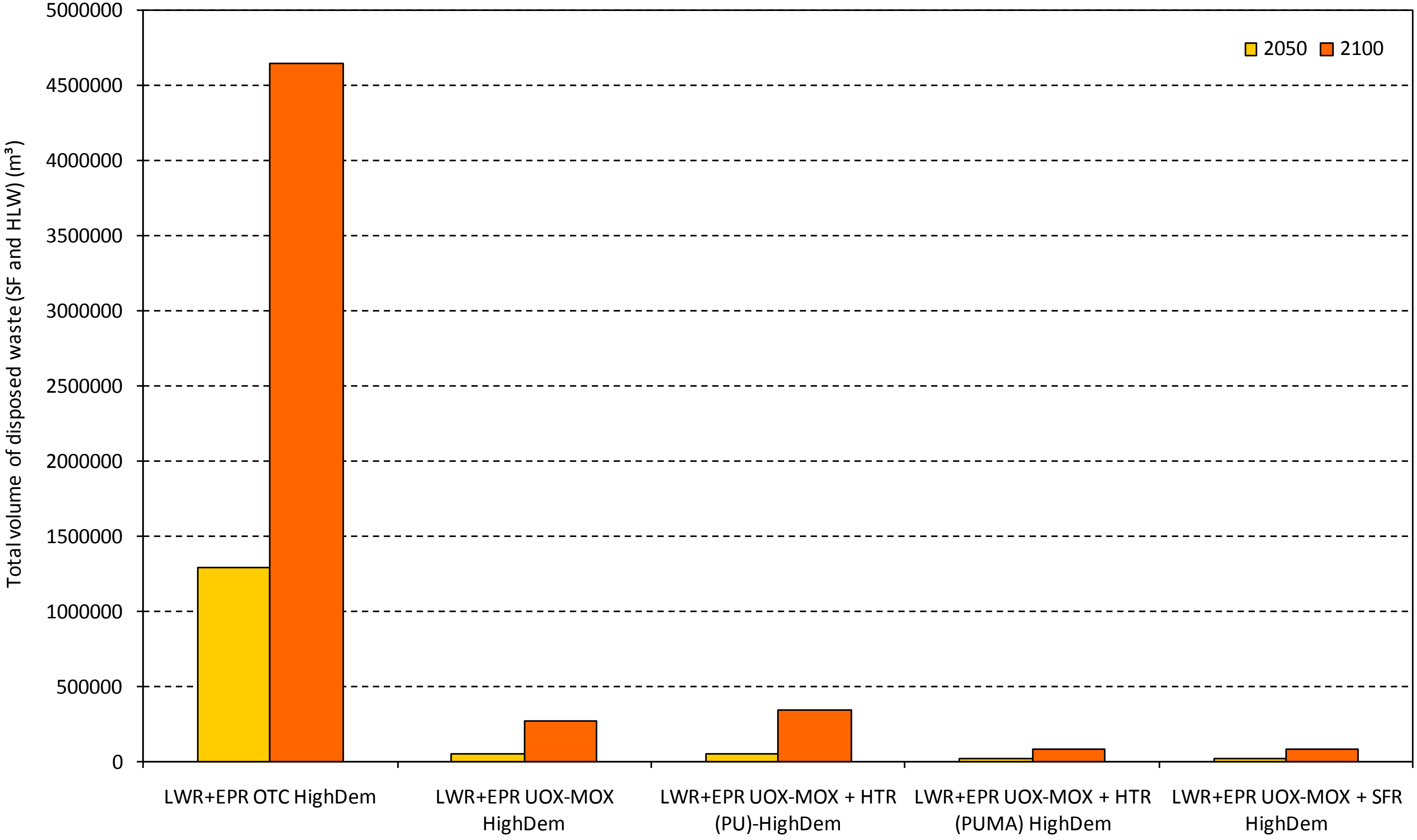
In-Pile TRU amounts for different scenarios



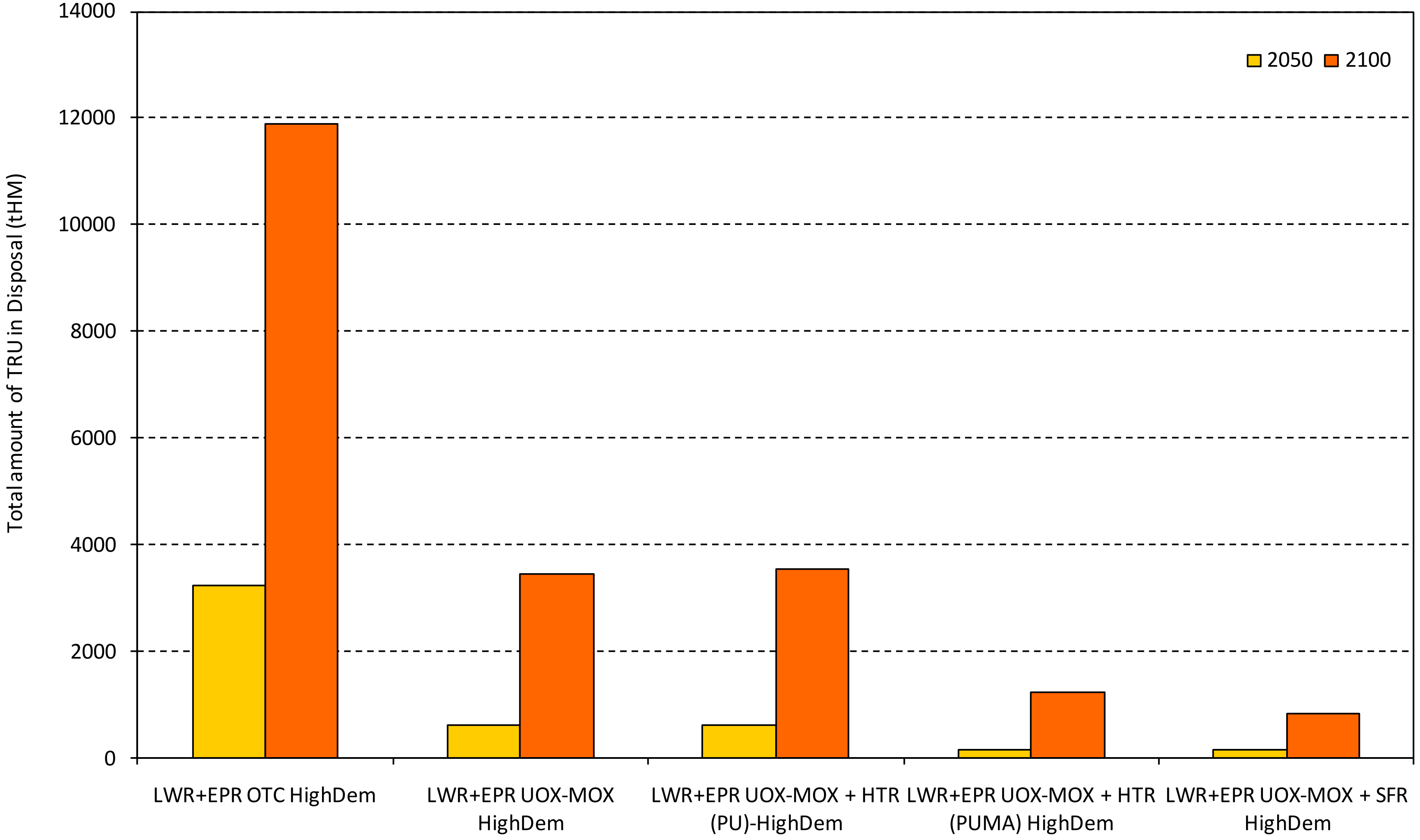
Comparison of Out-of-Pile TRU-amounts for the different scenarios



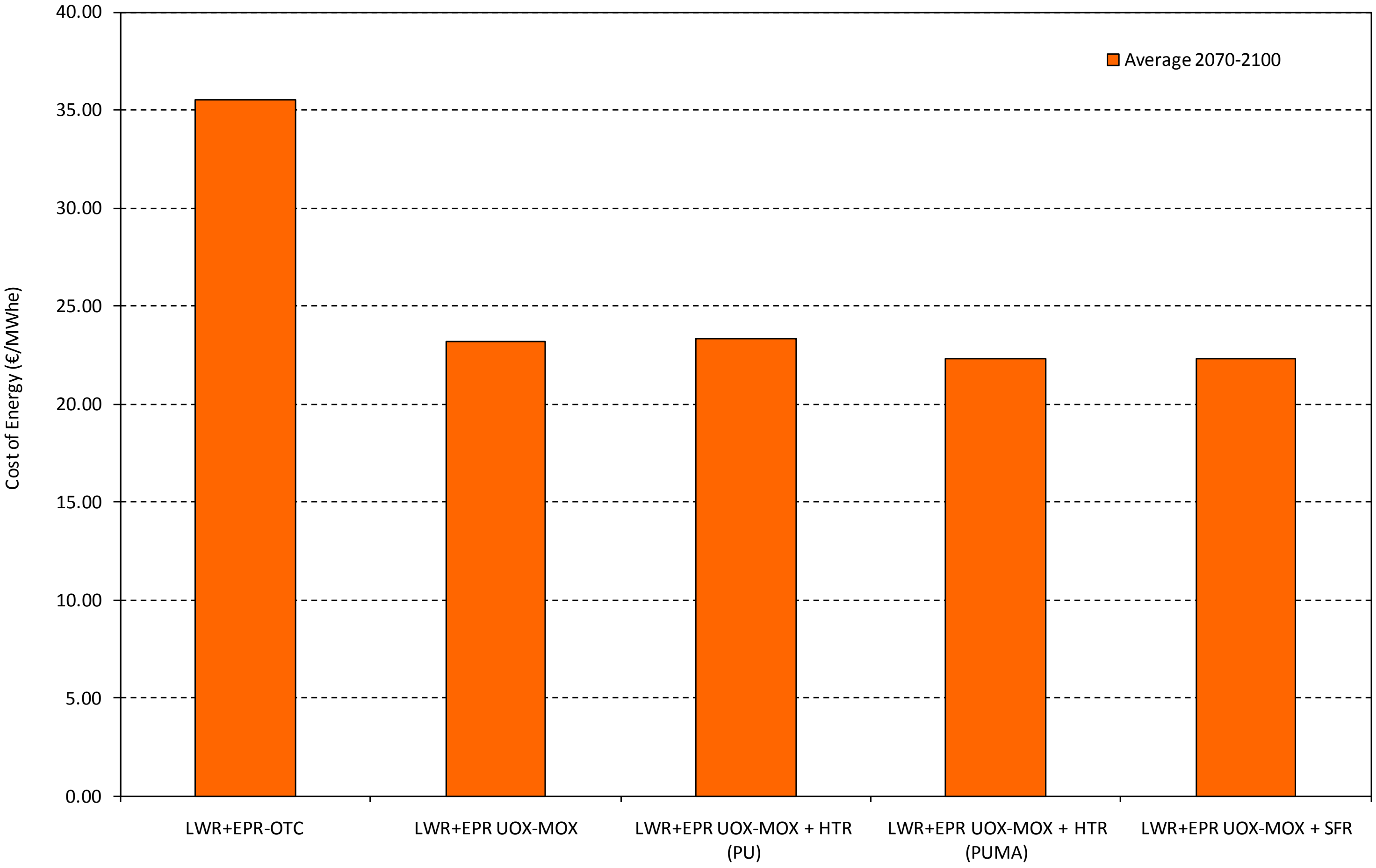
Comparison of disposed SF and HLW volumes for the different scenarios



TRU-amount in disposed SF and HLW for different scenarios



Averaged levelised energy generation costs for different scenarios



Conclusions

- Within the limitations and uncertainties of the scenario model it is clear that the introduction of Pu/MA HTRs may have a large impact on the amount of TRU in disposed spent fuel and HLW (close to FR scenario)
- Further investigations are highly recommended

WP4
Management, Communication, Knowledge Transfer

NRG (lead), LGI, IKE

Activities

- Define all project management activities of Coordinator and Project Management Office
- Contractual, administrative and financial management
- Work progress monitoring
- Definition of the project’s and consortium’s internal procedures
- Organisation, coordination and support for activities of dissemination of knowledge (contribution to RAPHAEL-PUMA-IAEA EUROCOURSE on V/HTR, Petten, December 2007)

Conclusion - Summary

- Main goal: to provide key elements for the use of contemporary and future (thermal) gas-cooled reactor designs as ***burner for plutonium and minor actinides***
- Core physics investigations aim at optimising CP fuel and reactor characteristics for Pu/MA loaded V/HTR core; promising results so far
- New (Pu/MA) CP designs are being explored
- Fuel performance models for Pu/MA HTR fuel are being developed
- Impact of introduction of Pu/MA-burning V/HTRs on entire fuel cycle and future nuclear energy mix have been investigated
- PUMA contributes to technological goals of Gen IV International Forum – addresses the sustainability issue of HTRs
- Many other issues concerning the use of Pu- and Pu/MA-bearing fuel in HTRs have been addressed in PUMA. See the project documents (listed in the PUMA Publishable Final Activity Report)

Follow-up...

A follow-up project for PUMA should at least address the following issues:

- Production and qualification (irradiation testing) of Pu/MA fuel (very high burn-up)
- Analysis tools have not been specifially validated for HTRs with Pu/MA fuel; validation should be based on experimental data (integral and separate effects)
- T/H-modelling should be 3-D instead of 2-D (R-Z): better identification/prediction of possible hot spots possible; MCNP-CFD
- Better modelling of pebble flow
- Further fuel cycle optimisation, possibly including Th
- Further development of fuel performance codes for Pu/MA fuel

QUO VADIS?

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