Materials for **I**nnovative **D**isposition by **A**dvanced **S**eparation: **MIDAS**

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The MIDAS Concept

Key objective

Improve the *safety* and sustainability of nuclear fuel cycle in the near term by *using advanced materials and separations* to achieve *innovative disposition and disposal of radioactive wastes.*

Relevance to Council Directive 2011/70/Euratom

39) Scientific research and technological development supported by *technical cooperation* between actors may *open horizons* to *improve* the *safe management* of spent fuel and radioactive waste, as well as contribute to *reducing the risk of the radiotoxicity of high-level waste*. ✓✓✓

Make it so that whatever I touch with my body, turns to yellow gold.

Ovid, Metamorphoses Book XI

Alignment with this meeting

IGD-TP considers that this WG should:

- Identify if very new issues coming from the future new reactors may arise without respect to the time frame; the view should be the largest possible at the beginning.
- Identify what could be the new materials, as well as the chemistry and waste packages.
- Develop, if necessary, new research taking into consideration new waste coming from new reactors.
- In addition, IGD-TP considers that some of the options may lead to re-analyze some issues of geological disposal.

Overall, the focus of this talk / proposed topic is synergistic with the above; the focus is on using separations technologies from advanced fuel cycles to improve sustainability with new Gen III reactors and safety of disposal in the immediate future. However, the proposed R&D would support and enable waste management beyond current Gen III.

✓✓

Objectives & boundary conditions

Objectives for proposed R&D by end of Horizon 2020

- Enhance sustainability of nuclear energy regeneration = fuel recycle
- Enhance safety of radioactive waste disposal = graded approach
- Enhance sustainable use of GDF resource = optimise use of footprint

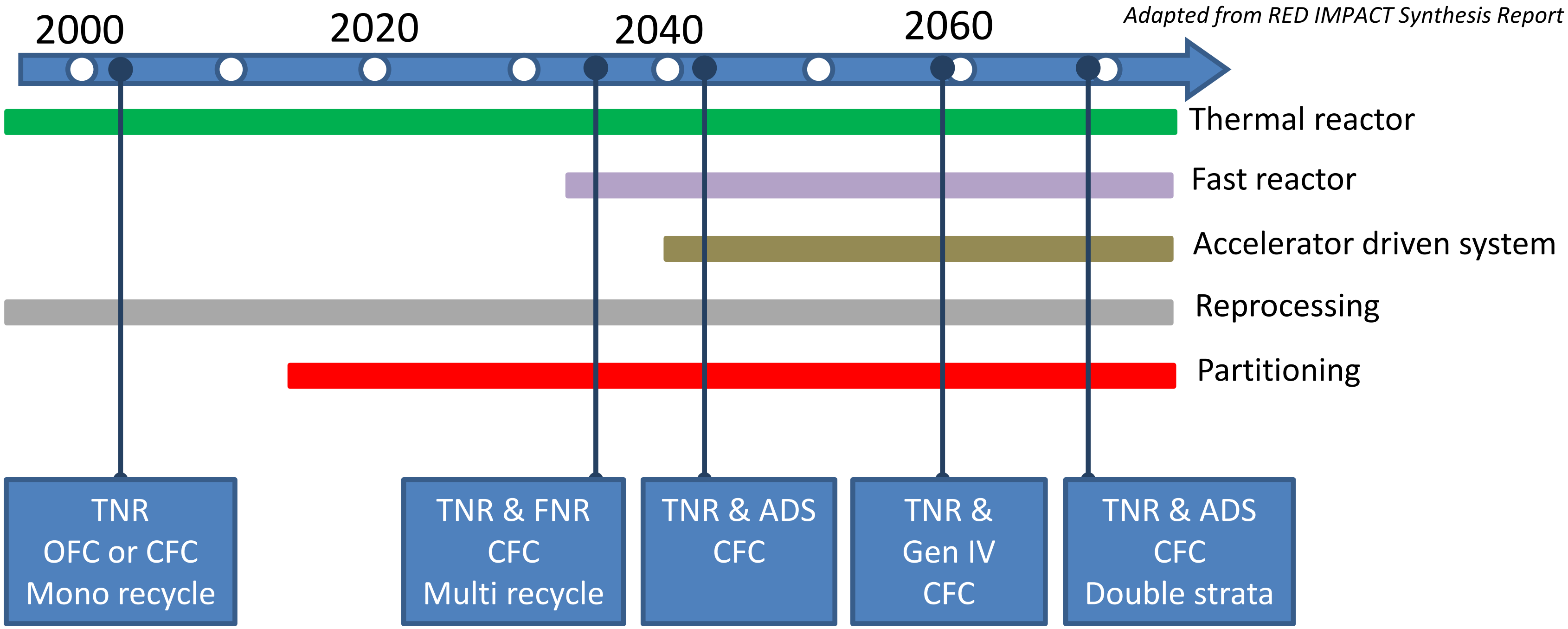
Key boundary conditions (assumptions)

- Work within the constraints of the European nuclear technology roadmap
- Leverage against / synergy with next generation strategy (FSR, P&T-ADS)
- Realistic opportunity of achieving improvement in current practice within 20-30 y

At least four BIG questions for new wasteform materials

1. What new materials could be deployed to achieve our objectives?
2. What are the grand challenges that need to be solved to enable deployment?
3. What new disposal strategies could be enabled?
4. What are the overall lifecyle benefits of the approach?

State of the art: FP6 RED IMPACT



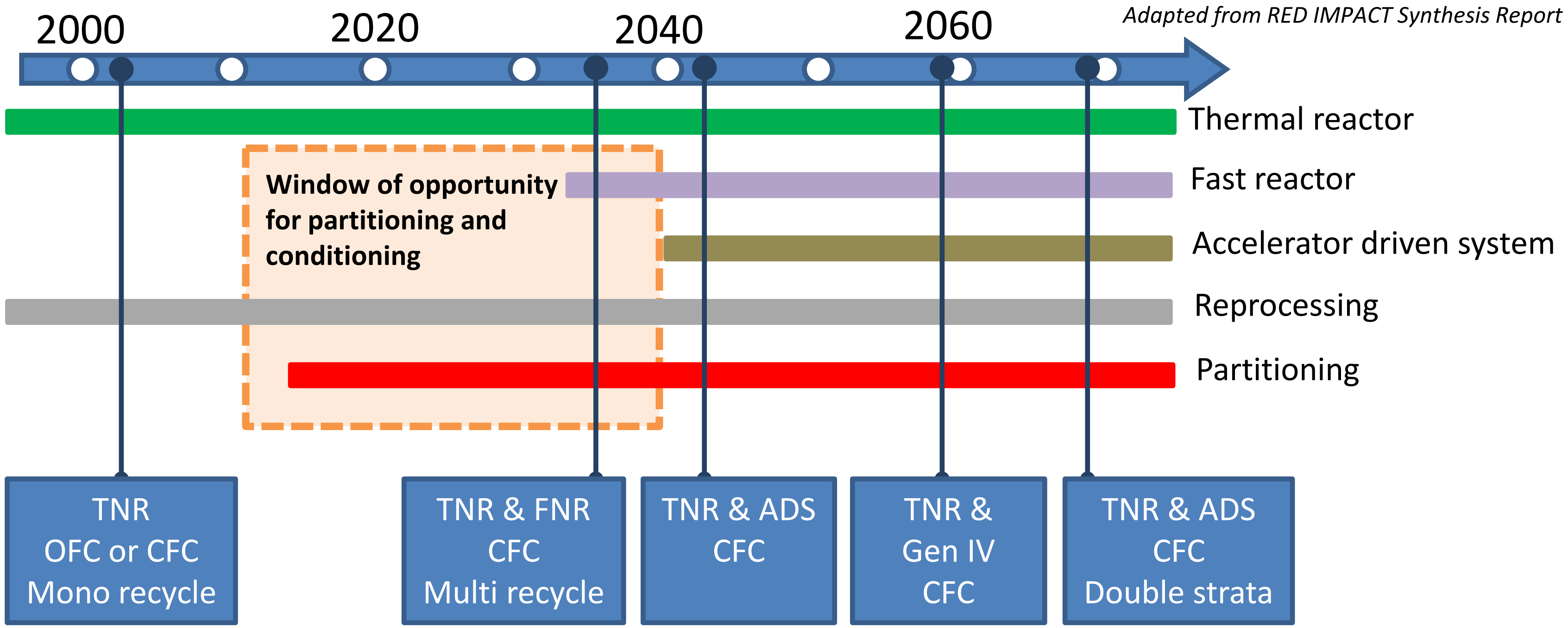
Adapted from RED IMPACT Synthesis Report

Within a 20-30 year timeframe it can be understood that the limiting technologies are:

- Thermal reactors for once through UOX and multi/mono UOX/MOX recycle
- PUREX reprocessing at industrial scale
- Availability of advanced partitioning flowsheets at pilot and industrial scale

How can advances improve sustainability and disposability without FNR or ADS?

State of the art: FP6 RED IMPACT



Adapted from RED IMPACT Synthesis Report

2000
2020
2040
2060

Thermal reactor

Fast reactor

Accelerator driven system

Reprocessing

Partitioning

Window of opportunity for partitioning and conditioning

TNR
OFC or CFC
Mono recycle

TNR & FNR
CFC
Multi recycle

TNR & ADS
CFC

TNR &
Gen IV
CFC

TNR & ADS
CFC
Double strata

<u>The MIDAS Window of Opportunity</u>

- Use emerging partitioning flowsheets for heat generating and long lived radionuclides
- Condition separated radionuclides in new advanced wasteforms
- Graded approach to interim storage and shallow, deep or very deep disposal
- Reduce heat burden and radiotoxicity consigned to deep disposal = sustainability
- Synergetic with long term strategy for FNR and ADS systems

State of the art: FP6 RED IMPACT

Impact of Cs/Sr separation & conditioning

- Separation of Cs and Sr results in HLW with very limited thermal out put and strong reduction in needed repository size.
- The long half life of ^{135}Cs requires Cs-waste to be disposed of in a repository according to current EU regulations.
- The feasibility of this option was not studied or assessed during the project.

Issue of long lived Minor Actinides

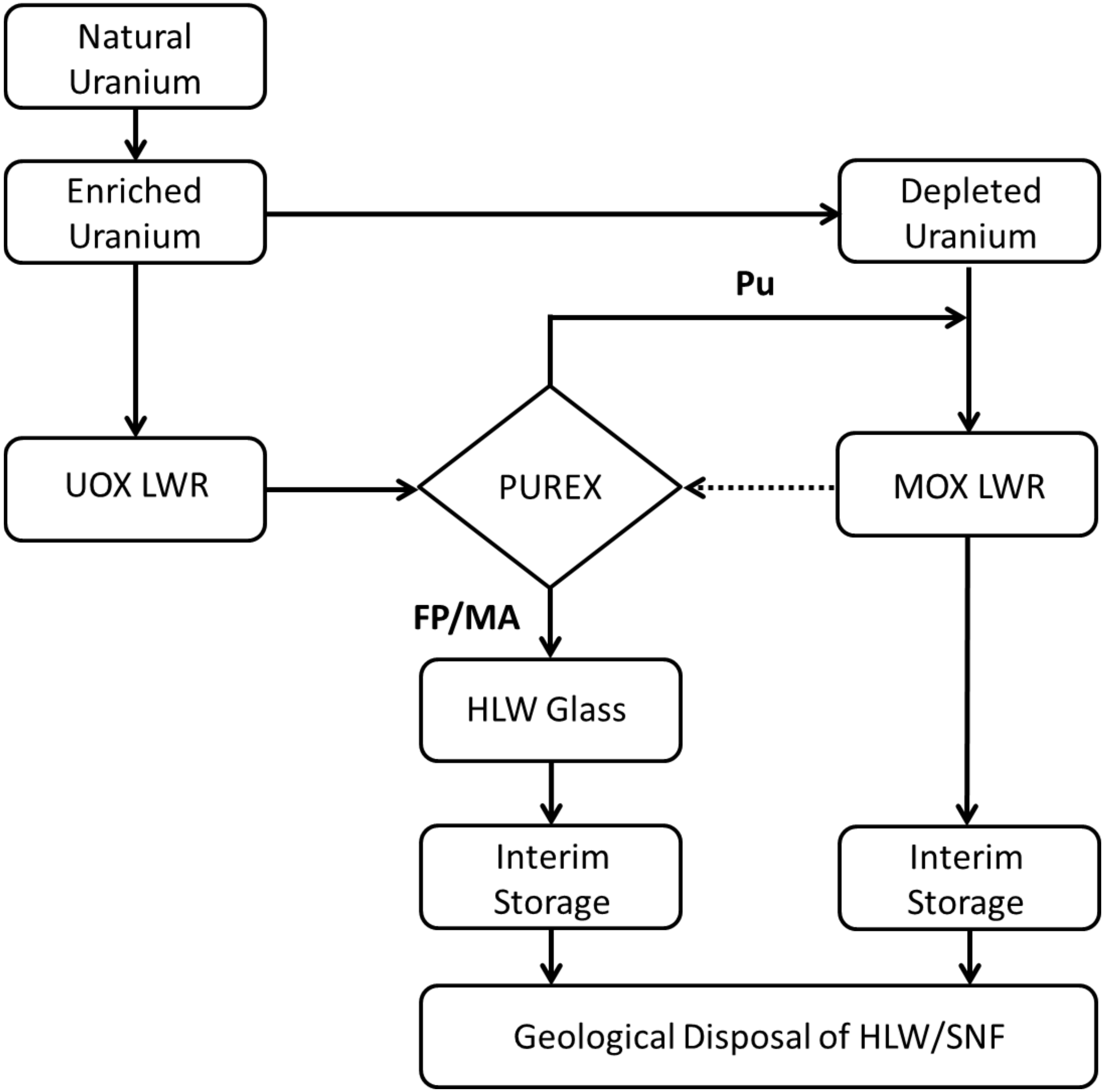
- Disposal in a repository will be required no matter what future scenario is chosen but public concern is one of the main impediments to continue and future use of nuclear energy.

Need for advanced wasteforms

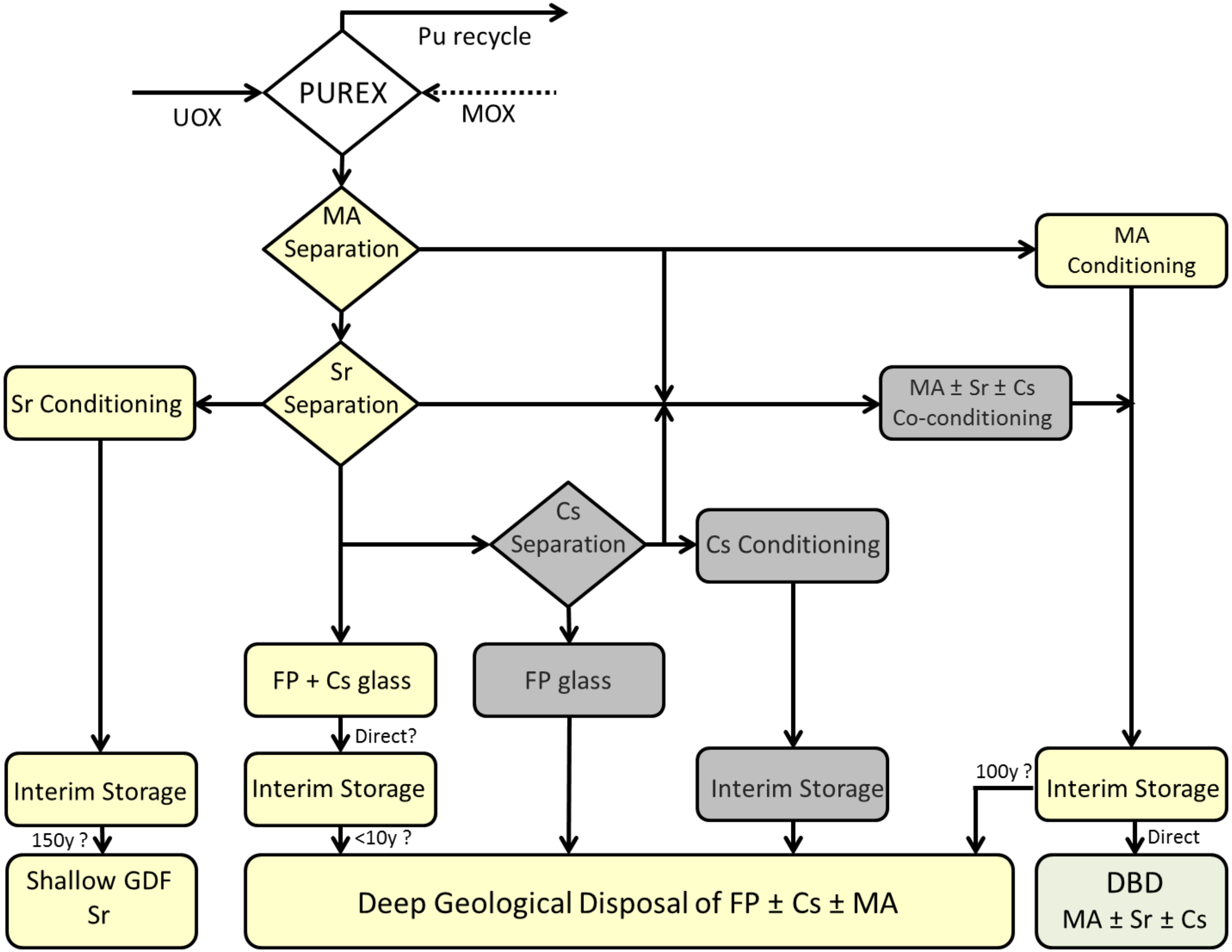
- Efforts should take account of the requirements to accommodate the waste streams emanating from advanced (minor actinide) partitioning with a view for specific conditioning.

Note strong synergy of proposal with RED IMPACT conclusions and perspectives.

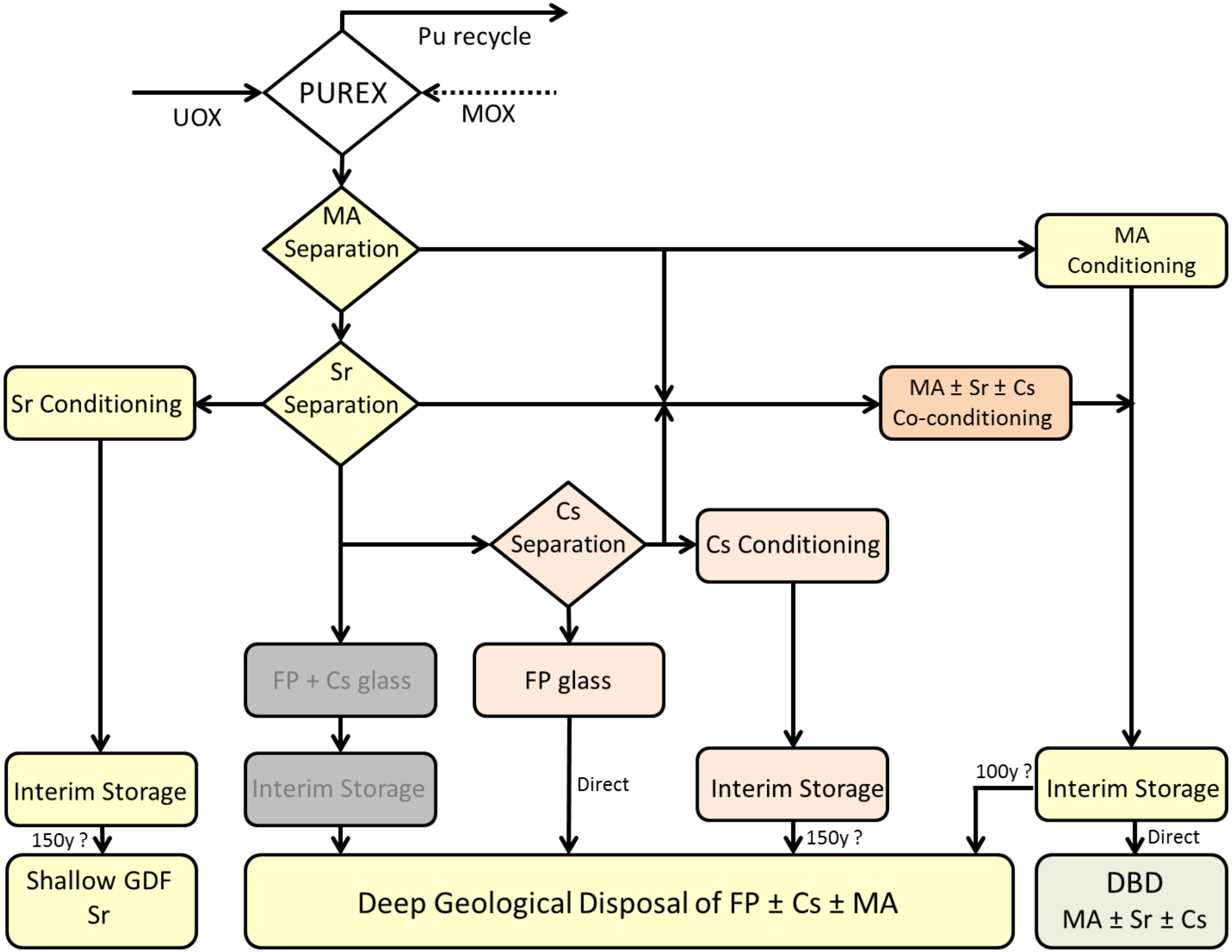
PUREX flowsheet



MIDAS flowsheet



MIDAS flowsheet



HT-DBD for MA wasteforms

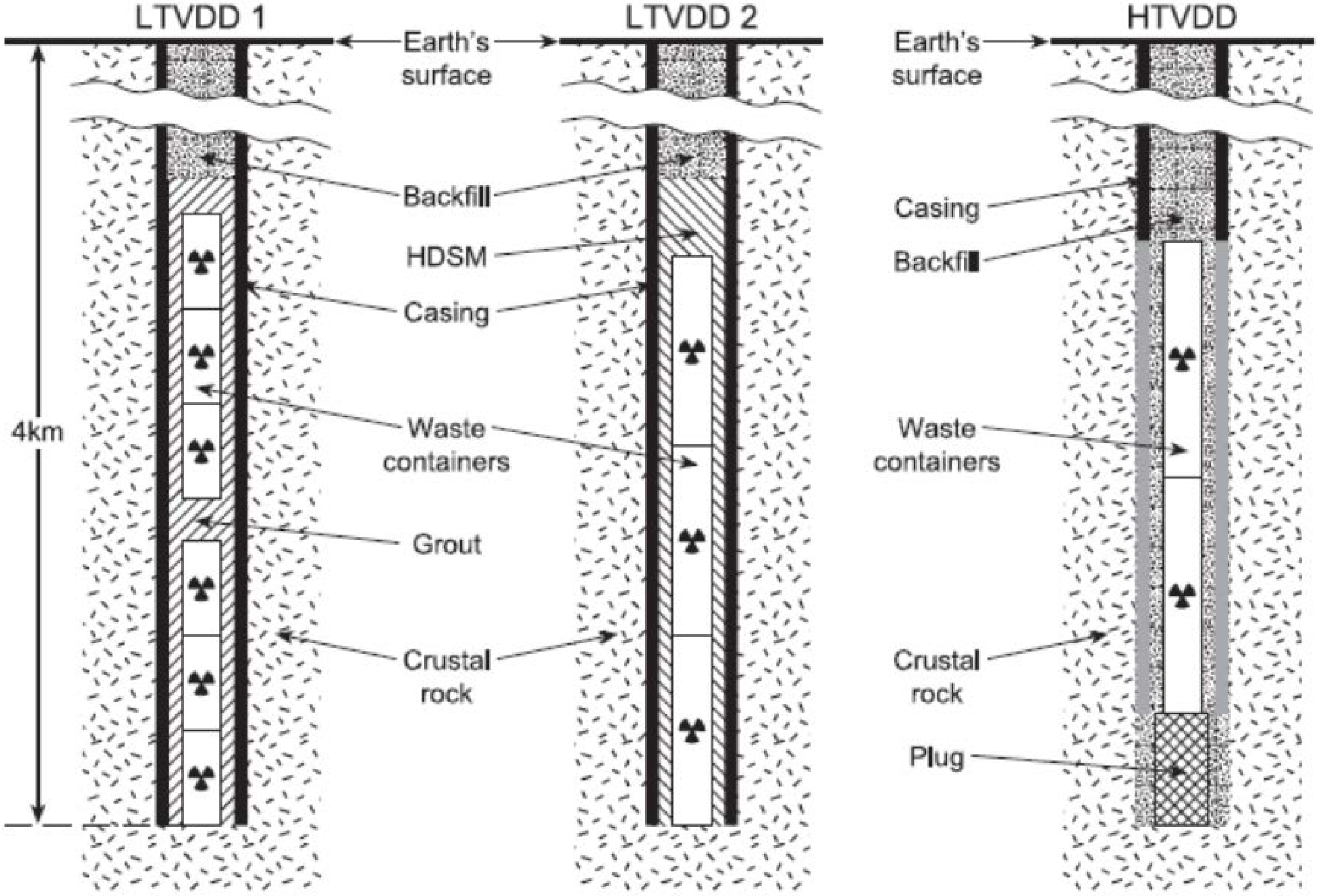


Figure 1. Three versions of deep borehole disposal (LTVDD = low-temperature very deep disposal, HTVDD = high-temperature very deep disposal, HDSM = high-density support matrix).

Partitioning status: RED IMPACT

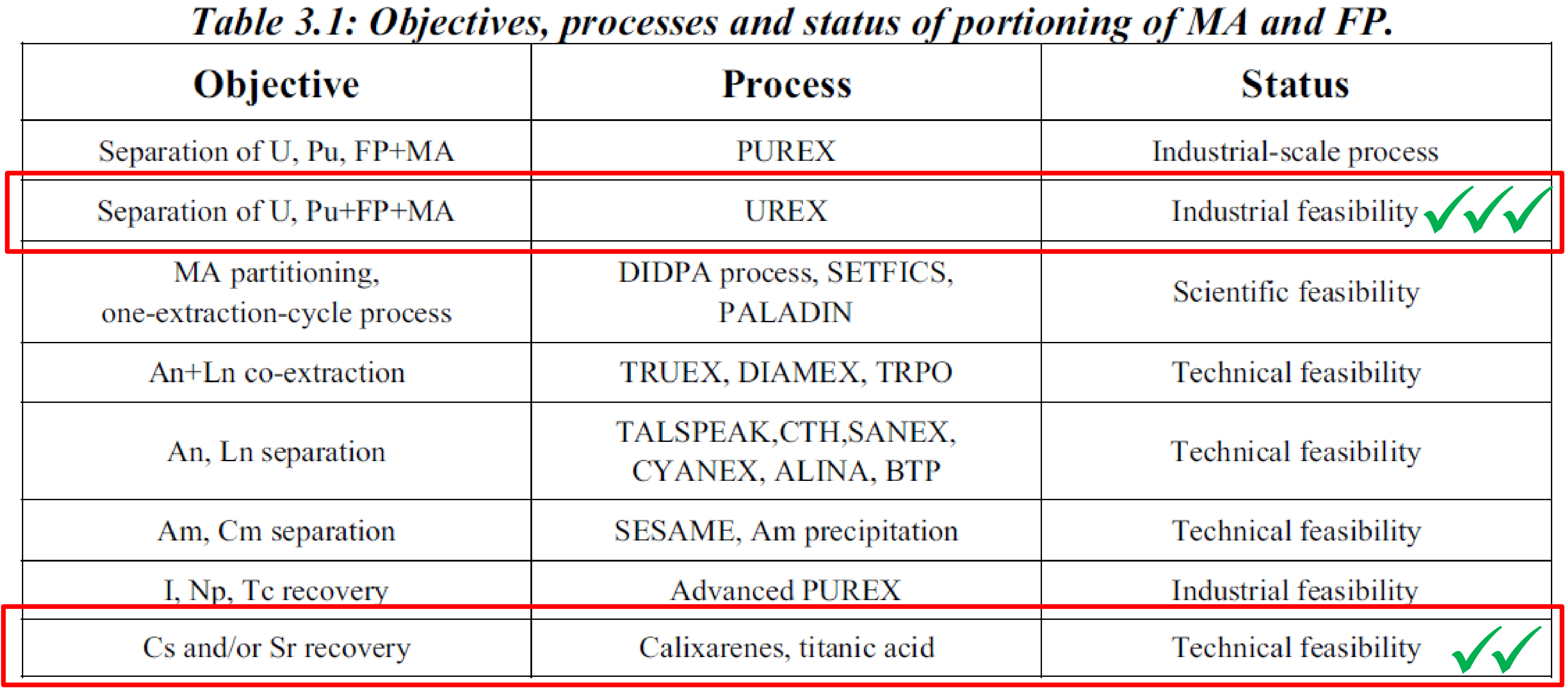


Table 3.1: Objectives, processes and status of portioning of MA and FP.

Objective	Process	Status	TRL
Separation of U, Pu, FP+MA	PUREX	Industrial-scale process	9
Separation of U, Pu+FP+MA	UREX	Industrial feasibility ✓✓✓	7/8
MA partitioning, one-extraction-cycle process	DIDPA process, SETFICS, PALADIN	Scientific feasibility	
An+Ln co-extraction	TRUEX, DIAMEX, TRPO	Technical feasibility	
An, Ln separation	TALSPEAK,CTH,SANEX, CYANEX, ALINA, BTP	Technical feasibility	
Am, Cm separation	SESAME, Am precipitation	Technical feasibility	
I, Np, Tc recovery	Advanced PUREX	Industrial feasibility	
Cs and/or Sr recovery	Calixarenes, titanic acid	Technical feasibility ✓✓	5/6

<u>The MIDAS Window of Opportunity</u>

- Flowsheets for MA separation are at technical or industrial scale feasibility
- Flowsheers for Cs/Sr separation are at technical scale feasibility

Hence, key scientific and technology requirement is for integrated conditioning matrices

General wasteform requirements

Requirements

- Adequate thermal conductivity for heat dissipation (could be assisted by Cermets?)
- Stable under long term self heating and $\beta.\gamma$ or α decay
- Accommodate impact of transmutation e.g. $^{90}Sr^{2+}$ to $^{90}Zr^{4+}$ or $^{241}Am^{3/4+}$ to $^{237}Np^{??+}$
- Acceptable trade-off between waste volume and heat output
- High durability of wasteforms if incorporating long lived species
- Manufacturing needs to be compatible with shielded cell
- Packaging design needs to be compatible with interim decay storage and final disposal

Glass wasteform

- Assumed by RED IMPACT and US AFCI
- No defined matrix: likely to be a silicate or phosphate glass
- Problem of T_g vs. heat output ; T_g increases with refractory nature, but so too does Cs volatility

Ceramic / glass-ceramic wasteform

- No defined matrix accepted: likely to be titanate or phosphate
- Basic knowledge of titanate and phosphate formulations and synthesis
- Problem of radiation induced amorphisation: micro-cracking, impact on long term performance

Potential Cs/Sr and MA wasteforms

Cs/Sr wasteforms

- Aluminosilicates: $CsAlSi_2O_6$, $SrAl_2Si_2O_8$
- Hollandite: $Ba_xCs_yM^{3+}{}_{2x+y}Ti_{8-2x-y}O_{16}$
- Perovskite: $SrTiO_3$
- Kosnarite: $NaZr_2(PO_4)_3$
- Others, e.g. $Cs_2ZrSi_6O_{15}$

MA wasteforms

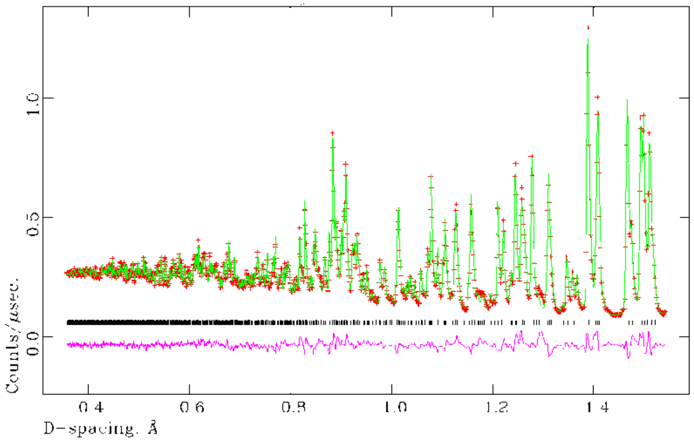
- Zirconia / pyrochlore / zirconolite: ZrO_2, $Ln_2Ti_2O_7$, $Ln_2Zr_2O_7$, $CaZrTi_2O_7$
- Monazite: $LnPO_4$
- Perovskite: $(Sr,Act)(Ti,M)O_{3-x}$
- Garnet: $(Ca,Ln,Act)_3ZrFe_4O_{12}$
- Britholite: $Ca_4Ln_6(SiO_4)_6O$
- LaBS / Loeffler glass, e.g. 38% Ln_2O_3, 28% SiO_2, 17% Al_2O_3, 3% B_2O_3, 14% other

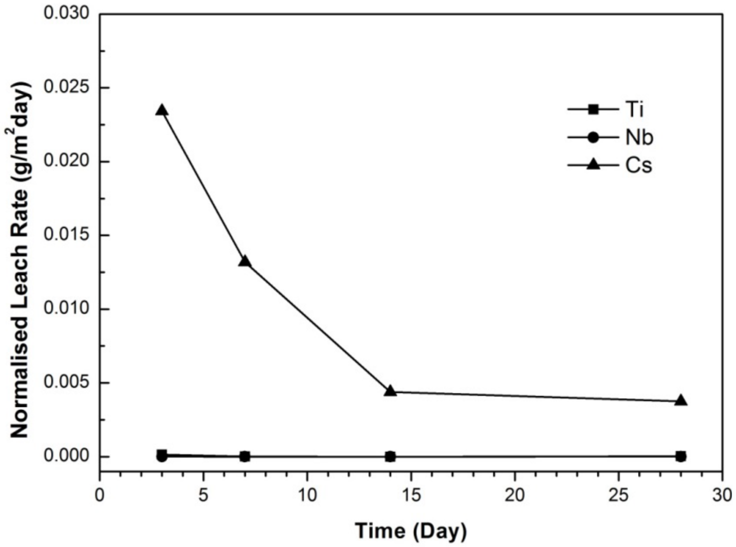
Grand challenges for Cs/Sr & MA wasteforms

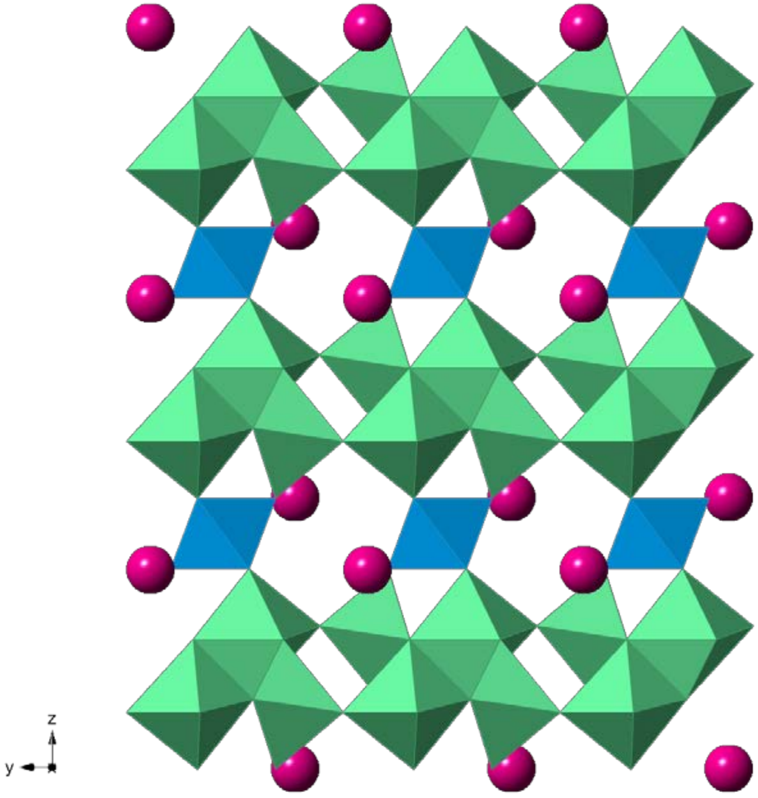
- Optimisation of separations flowsheet, in particular effects of radiolysis and heat output on separation factor and solvent / extractant recycle .
- Optimisation of ceramic / glass formulations: for waste loading / volume, and thermal output, radiation stability and durability in storage and disposal systems
- Theoretical understanding and mechanistic models of transmutation in wasteforms and crystalline to amorphous phase transition by α-decay
- Need mechanistic models of effect of prolonged self heating and high centreline temperature: devitrification of glasses; solid state diffusion; micro-cracking?
- Need integrated mechanistic models of chemical and microstructural impact of transmutation and amorphisation on dissolution behaviour.
- Need for an integrated strategy of staged wet and dry interim storage for heat generating wasteforms.
- Need for new disposal and EBS concepts: shallow GDF for Sr; dedicated galleries in deep GDF for Cs and MA±Cs/Sr; deep boreholes for MA.
- Need to demonstrate manufacturing route consistent with remote handling constraints.

Example from UoS /UoB collaboration

Synthesis of $Cs_2TiNb_2O_{18}$: an new conditioning matrix for radio-cesium



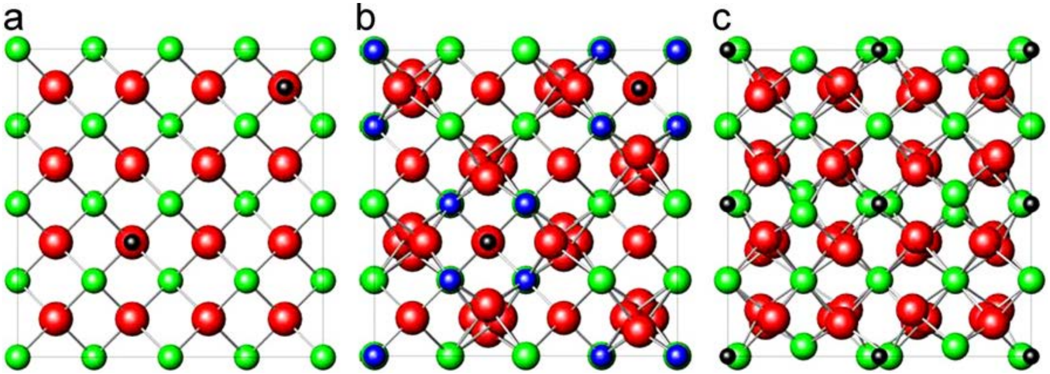




Example from UoS work

Investigation of Ce (Pu) substitution in $Gd_2Zr_2O_7$ pyrochlore ceramic

Key question: is Ce accommodated by nano-scale phase separation?



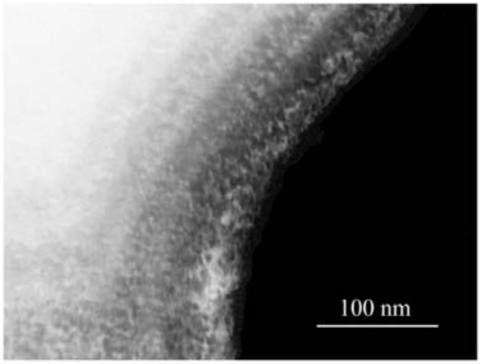
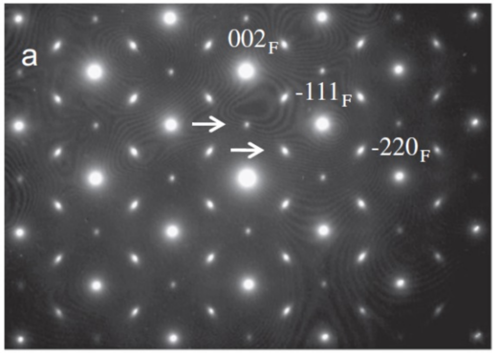


Fig. 7. Satellite dark field image of $Gd_2(Zr_{1.5}Ce_{0.5})O_7$ obtained using $G_F \pm \frac{1}{2}(1\,1\,1)^*$ reflection.



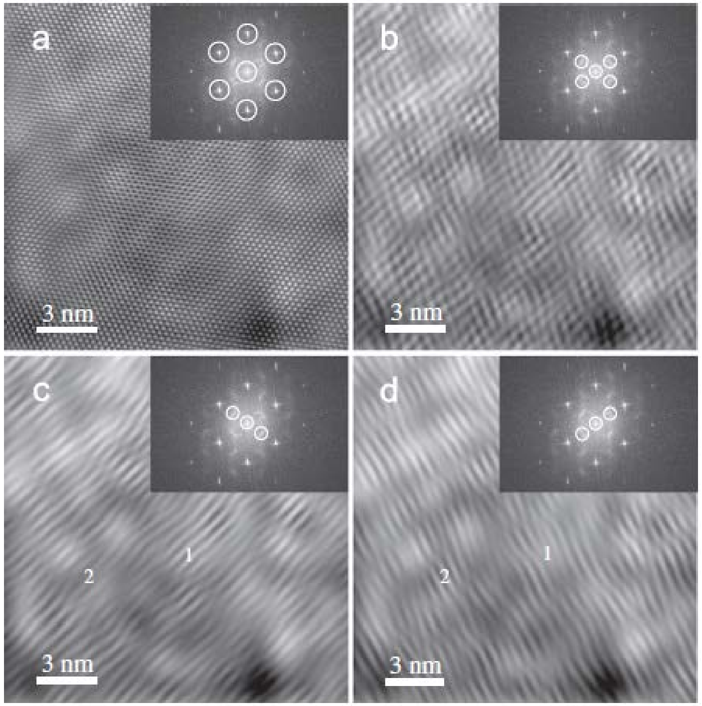


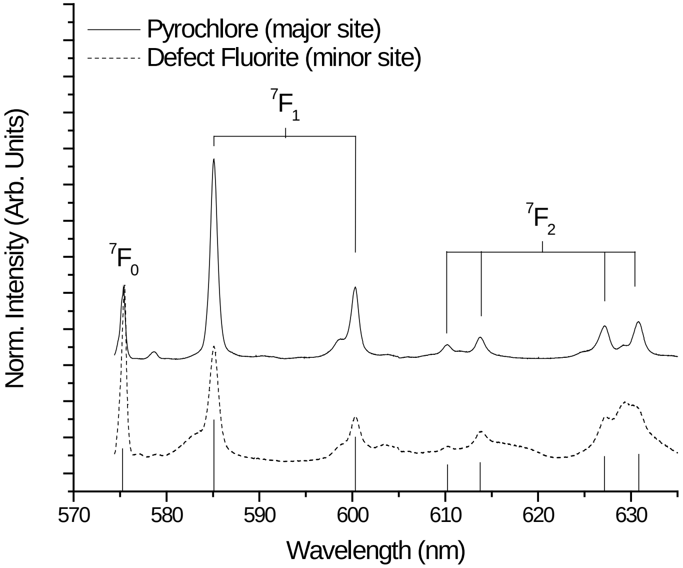
Fig. 9. Processed HREM images using Fourier masks positioned as shown in the power spectrum insets. (a) Processed image using central transmitted beam and the fluorite sub-cell reflections. (b) Processed image using the central transmitted beam and the diffuse intensity at all the $G_F \pm \frac{1}{2}(1\,1\,1)^*$ positions. (c and d) Processed images using the central transmitted beam and the diffuse intensity in one of the two orthogonal sets of $G_F \pm \frac{1}{2}(1\,1\,1)^*$ positions.

Example from JÜLICH

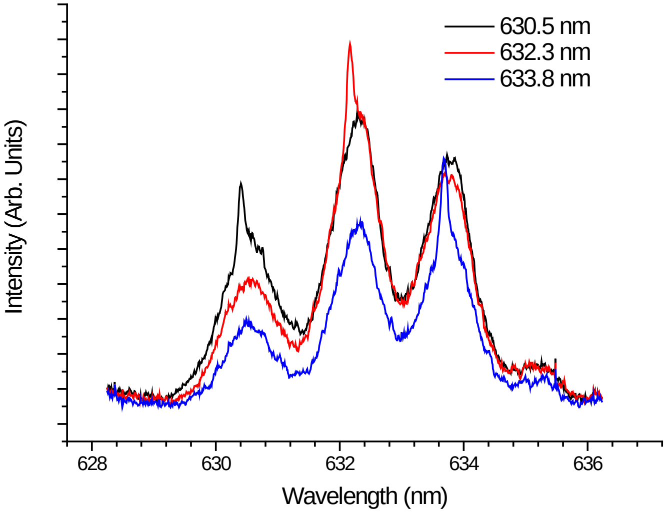
Eu and Cm doped $La_2Zr_2O_7$ with pyrochlore or defect fluorite crystal structure

- Identification of structural sites of Ln^{3+}/An^{3+} within the crystal structures

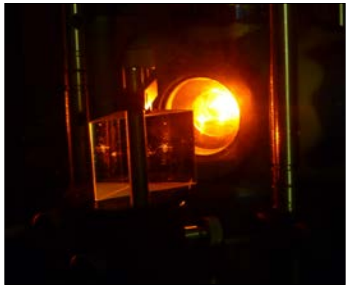
Key question: Understand the stability of pyrochlore as potential wasteform (e.g. response to radiation damage)



Emission spectra after direct excitation of Eu to the $^5D_0$ band



Emission spectra after direct excitation of Cm to the $^6D_{7/2}$ band



Potential project outline

WP	Research focus
1	**Optimisation and integration of advanced separations** *Modelling for industrial scale up; impact of radiolysis and (radiogenic) heating on efficiency and solvent / extractant recycle.*
2	**Solving grand challenges for Cs/Sr and MA wasteforms** *Focus on wasteform design and formulation for Sr, Cs, MA±Cs±Sr; transmutation & radiation damage effects; thermal conductivity; trade off of volume, heat output, radiation effects, and durability.*
3	**Manufacturing technology readiness** *Cold full scale production of optimised wasteforms, concept design study for integration into hot cell environment.*
4	**Integrated storage and disposal strategies** *Concept designs for decay-storage; impact of new wasteforms on existing disposal concepts (footprint, dose models, etc); concept designs for alternative disposal strategies (shallow GDF for Sr; DBD for MA)*
5	**MCDA of whole lifecycle benefits** *Analysis of cost, safety, manufacture, timescale, and future dose of parameterised scenarios of deployment*

Alignment with SNETP strategy

From SNETP Strategic Research & Innovation Agenda 2013

To optimise HLW management, research should focus on minimising several parameters of the HLW:

- The mass and volume of conditioned waste to be disposed of
- The long term “radiotoxic inventory” to be disposed of
- The effective “lifetime” of conditioned waste
- The heat generation of conditioned waste as function of time…. This parameter strongly affects the GDF capacity
- The “long-term radiological impact”, that is the calculated biological effect on living species of possible radioactive releases into the biosphere once part of radionuclides (or their radioactive daughters) has reached the surface

✓✓✓

Alignment with IGDTP strategy

From **IGDTP** SIRA 2011 and Deployment Plan 2011-16

Key topic areas defined:

1. Safety case
2. Waste forms and their behaviour
3. Technical feasibility and long-term performance of repository components
4. Development strategy of the repository
5. Safety of construction and operations
6. Monitoring
7. Governance and Stakeholder involvement

IGDTP SIRA & DP appears silent on relevance of new wasteforms and interaction with SNETP SIRA! Threat or opportunity?

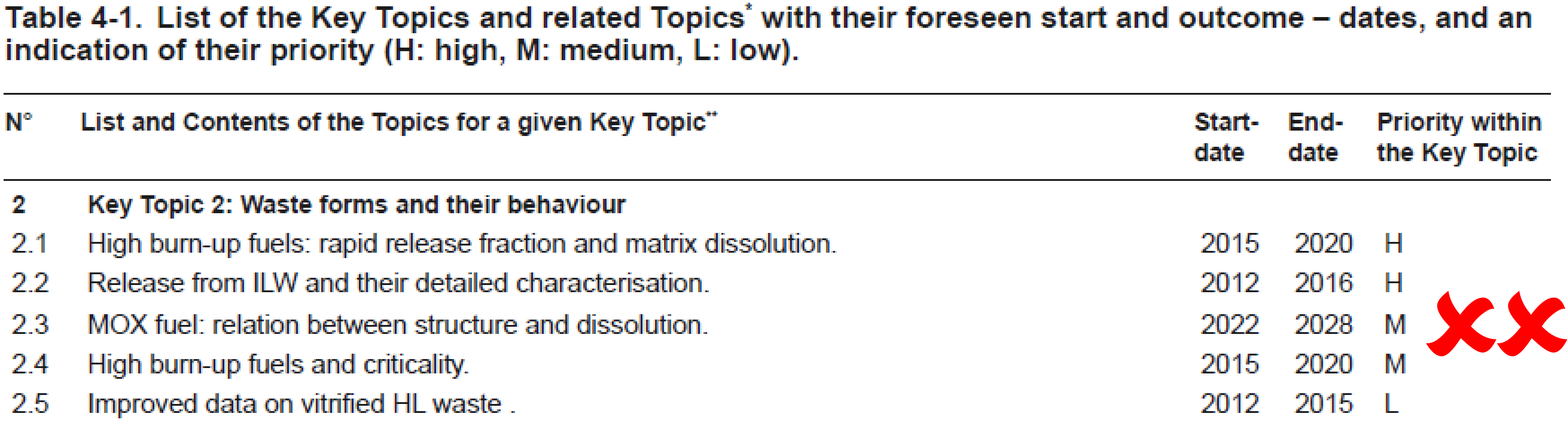
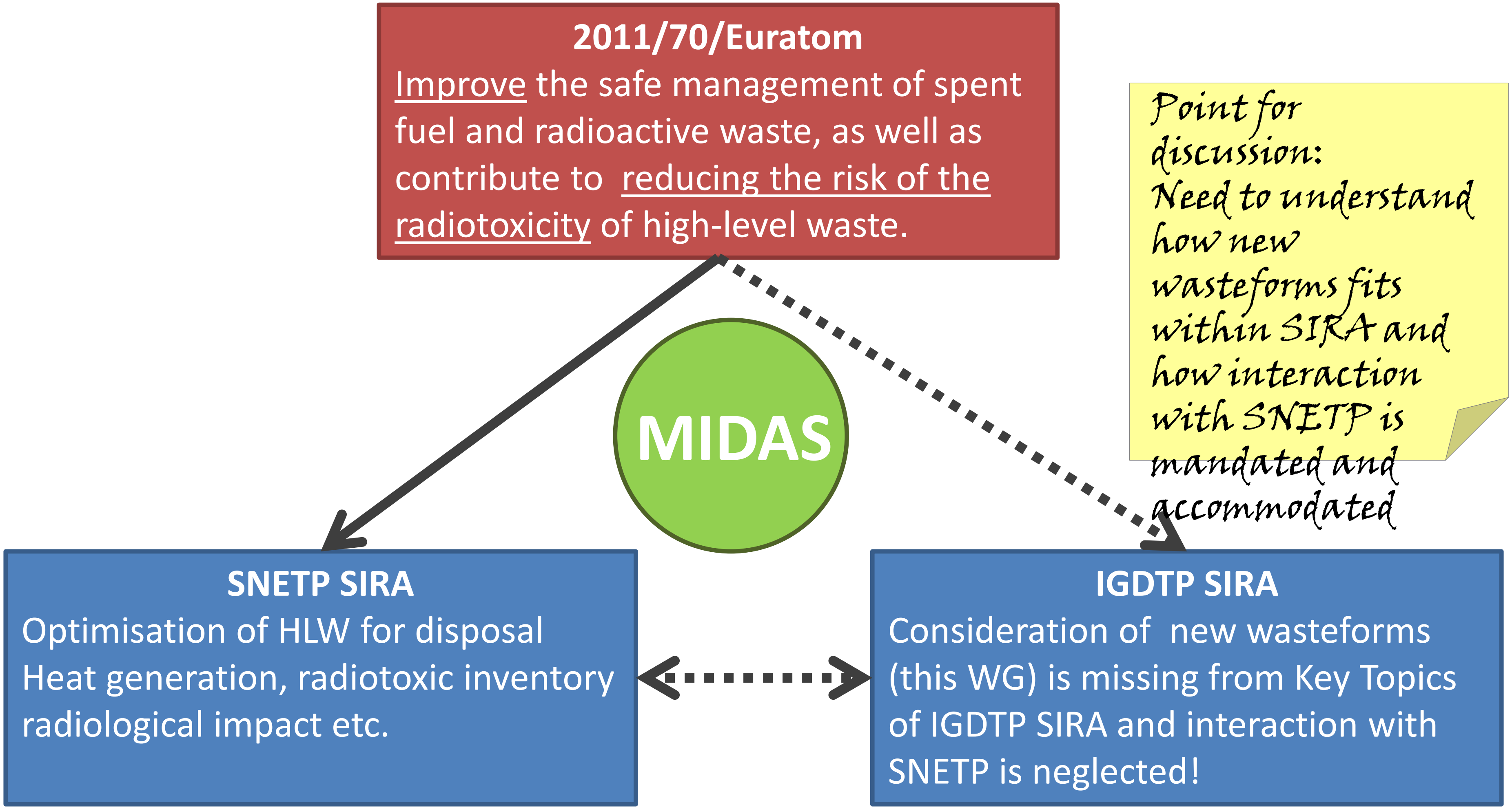


Table 4-1. List of the Key Topics and related Topics[*] with their foreseen start and outcome – dates, and an indication of their priority (H: high, M: medium, L: low).

N°	List and Contents of the Topics for a given Key Topic[**]	Start-date	End-date	Priority within the Key Topic
2	**Key Topic 2: Waste forms and their behaviour**			
2.1	High burn-up fuels: rapid release fraction and matrix dissolution.	2015	2020	H
2.2	Release from ILW and their detailed characterisation.	2012	2016	H
2.3	MOX fuel: relation between structure and dissolution.	2022	2028	M
2.4	High burn-up fuels and criticality.	2015	2020	M
2.5	Improved data on vitrified HL waste .	2012	2015	L

✗✗✗

Possible strategy disconnect?



2011/70/Euratom

Improve the safe management of spent fuel and radioactive waste, as well as contribute to reducing the risk of the radiotoxicity of high-level waste.

MIDAS

SNETP SIRA

Optimisation of HLW for disposal
Heat generation, radiotoxic inventory
radiological impact etc.

IGDTP SIRA

Consideration of new wasteforms (this WG) is missing from Key Topics of IGDTP SIRA and interaction with SNETP is neglected!

Point for discussion:
Need to understand how new wasteforms fits within SIRA and how interaction with SNETP is mandated and accommodated