

ROLE OF MINOR ACTINIDES MINIMIZATION TECHNOLOGIES AND APPROACHES IN ADDRESSING ECO-FRIENDLY ADVANCED NUCLEAR FUEL CYCLES

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Outline of the presentation

- IAEA Programme of Work
- Partitioning & Transmutation potentials
- Advanced fuel cycles MA technologies
- Fuel cycle approaches, LMFRFC
- Thorium fuel cycle, Inert Matrix Fuel includes coated particle
- MA-Fuel / databank
- Link between process losses and environmental impact
- Role of Pyro-chemical processes in AFC
- Concluding Remarks







Department of Nuclear Energy

Division of Nuclear Fuel cycle and Waste technology





Genesis of the problem Partitioning and Transmutation to deal with nuclear waste

- Reduction of long-term radiotoxicity
- Determination of a period to terminate the disposal site (From over million years to less than ~700 years)
- Reduction of repository space requirements
- Increased resource utilization
- Scientific, technical and economical feasibilities
- Reduce Proliferation concern



CEA Program on Partitioning & Transmutation, D. Warin and H. Safa, *International Meeting on Accelerator Driven Transmutation System Technologies, University of Nevada, Las Vegas, April 29, 2003*



IAEA cooperations in P&T Activities

► Evaluation of actinide Partitioning and Transmutation (1982) \succ Use of fast reactors for actinide transmutation (1993) \succ Feasibility of separation of noble metals from HLLW (1989) Transmutation of separated Pu ► Review of nuclear data for ADS related R&D (1998) Overview of development of accelerator-driven sub-critical reactors (ADS) for transmutation of waste (1998) ➤Utilization of thorium based fuel-cycle for ADS (1998) \succ Feasibility of hybrid reactor concepts (1999) \rightarrow Non-proliferation and environmental aspects of P&T (2001) Emerging nuclear energy and transmutation systems: Core physics and engineering aspects (2003)





Advanced Fuel cycle

Fuel Cycle that incorporate P&T to burn TRU elements is called Advanced Fuel Cycle

- 1. Separation of TRUs and LLFPs
- 2. Fabrication of TRU-rich fuel and targets
- 3. Transmutation in Thermal &/or Fast reactors or ADS



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Advanced Fuel Cycle consists

- a) Reprocessing of LWR-UO₂ fuel
- b) Separation of minor actinides from HLLW resulting from LWR-UO₂ reprocessing
- c) Fabrication of MA targets for heterogeneous irradiation in LWRs
- d) Quantitative recycling of U and Pu into LWR-Mixed Oxide (MOX) fuel
- e) Reprocessing of spent LWR-MOX fuel in adequate facilities (higher Pu inventory)
- f) Separation of MAs from HLLW & conditioning of individual elements MAs
- g) Fabrication of FR -fuel (MOX, -metal or -nitride) with limited MA content
- h) Irradiation in FRs or dedicated hybrid facilities to very high burnup
- i) Reprocessing of spent FR-fuel in specially (aqueous and non-aqueous)
- j) Quantitative separation of all TRUs from the spent FR (and /or ADS) fuel processing during multiple recycling
- k) Multiple recycling of FR-MOX fuel with major TRU content until significant depletion
- 1) Separation of certain fission products with long half-lives if required for the disposal step

Source: P&T as a Waste Management Option in a Future Nuclear Era, L.H. Baestslé, in Proc. Workshop on Hybrid Nuclear Systems for Energy Production, Utilisation of Actinides & Transmutation of Long-lived Radioactive Waste, IAEA, Sep 3-7, 2001, ICTP, Trieste, Italy, and IAEA, Vienna, Austria, 2001, IAEA/SMR/1326-3 ARWIF Workshop 16-18 Feb 2005

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Source: Accelerator-driven Systems (ADS) and Fast Reactors (FR) in Advanced Nuclear Fuel Cycles, A Comparative Study, (2002) NUCLEAR ENERGY AGENCY ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT









Schematic ATW waste treatment flowsheet.



Fabrication experience of LMFR fuels in Laboratory(L), Pilot(P) & Commercial(C) scales

SI. No.	Country	Oxide UO ₂ ,(U,Pu)O ₂	Carbide UC, (U,Pu)C	Nitride UN, (U,Pu)N	U-Fs &U- Pu-Zr
1.	USA	Р	L	L	Р
2.	Russia	С	L	L	
3.	France	С	L	L	
4.	India	L	Р	L	
5.	Japan	Р	L	L	
6.	UK	С	L		
7.	Germany	С	L		

Fabrication R&D on LMFR fuels for Minor actinide based fuels

SI. No.	Country	Oxide	Advanced fuel forms	Nitride	Metallic
1.	USA	Yes	Yes		Yes
2.	Russia	Yes	Yes	Yes	
3.	France	Yes	Yes		
4.	Japan	Yes		Yes	Yes
5.	EU	Yes	Yes		Yes



LMFR-System Rests on Environmentally Benign Nuclear Fuel Cycle

Reduced radio-toxic waste inventories as well as longevity (long-term radio-toxicity)
Reduced containment duration

- •Less stringent repository requirements
- Increased resource utilization

Some Potential Challenges in Addressing Environmentally Benign LMR-FR Fuel Cycles

- Development of transuranic-based fuel and fuel cycles
- Reprocessing of such fuels, in combination with the immobilization of separated non-reusable radiotoxic nuclides and noble metals for ultimate disposal
- Suitable matrixes for ¹⁴C and ¹⁵N should be developed for the realization of advanced fuels such as coated particle or nitride fuel etc.,
- Methods to recycle and reuse fission products should be addressed
- To increase the capacity of repository by separating the heat producing radio-nuclides and prepare these for cooling storage
- Operational exposure: minimize workers' exposure-limits for such fuel fabrication and spent fuel treatment.





Technical Meeting on

"Current Status and Future Prospects of Liquid Metal Cooled Reactor Fuel Cycle and fuels" (TM-LMFR-2005) 3-7 Oct 2005*

at

Central Institute of Improvement of Qualification (TSIPK), Obninsk, Russian Federation

with cooperation from Institute of Physics and Power Engineering (IPPE) Obninsk, Russian Federation

* Subject to clearances and approvals



Provisional programme

- Role of LMFRs in sustainable nuclear energy development
- General LMFRs developments / new concepts
- Current status of LMFR fuel cycles
- Economics of LMFBR fuel cycles and its comparison with the other fast reactor fuel cycles
- Current status of liquid metal-cooled reactors fuels
- Advanced fuel fabrication, design including conventional one/new concepts
- Development of fuel fabrication technology including QA and QC
- Irradiation testing, fuel performance in normal and transient conditions and fuel modelling
- Existing and planned facilities and experimental programs
- Advances in materials development for fuel cycle application
- Trends in innovative fuel cycle concepts including partitioning and transmutation pertinent to LMFR fuel cycle development specially encompassing the subject areas:
 - Theory, Design, Development, irradiation testing and performance
 - Status and innovations in partitioning as well as transmutation methods Waste reduction, proliferation-resistance and environmental improvement through LMFR fuel cycles
 - Challenges and R&D needs in LMFR fuel cycle
 - o Cross-cutting and infrastructure issues
- Knowledge conservation on LMR fuel cycle
- Prospects for international collaboration and co-ordination of R&D activities
- IAEA's role in meeting the needs for information exchange and collaborative R&D.





Thorium fuel cycle

M. Lung, O. Gremm / Nuclear Engineering and Design 180 (1998) 133-146

MINOR ACTINIDES	DIVERSE FUEL CYCLES			
	U5 + U8 (Reference cycle) (grams per ton HM)	U5 + Th2 (%)	U3+U8 (%)	U3 + Th2 (%)
Np-237	360 *	92	20	1
	900 **	107	13	3
Am	160 *	0.04	106	6.3 10 ⁻⁵
(241 + 242 + 243)	470 **	0.28	117	1.8 10 ⁻³
Cm	36	0.01	111	1.68 10 ⁻⁵
(243 to 246)	220	0.14	132	6.37 10 ⁻⁴

* Discharge burn-up : 30 GW d/t ** Discharge burn-up : 60 GW d/t

Relative production of minor actinides in diverse cycles

Thorium fuel cycle

Very large thorium resources could be utilized for nuclear energy generation apart from increasing efficiency of U-resource utilization

- Neutron yields of ²³³U in the thermal and epithermal regions are higher than those of ²³⁹Pu and also , ²³²Th is a better 'fertile' material than ²³⁸U in thermal reactors
- Improved operating margins due to better thermo-physical properties of ThO₂ fuel even at high burnup, compared to those of UO₂ fuel
- Potential for fuel cycle cost reduction, the reduction in ²³⁵U enrichment requirements
- Reducing long-lived radioactive waste inventories by diminishing the production of plutonium and minor actinides
- Th-based fuels and fuel cycles have intrinsic proliferation-resistance due to the formation of ²³²U via (n,2n) reactions with ²³²Th, ²³³Pa and ²³³U. The half-life of ²³²U is only 73.6 years and the daughter products have very short half-life and some like ²¹²Bi and ²⁰⁸TI emit strong gamma radiations



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Name and Country	Туре	Power	Fuel	O peration period
AVR, Germany	H T G R E x p e r i m e n t a l (P e b b l e b e d r e a c t o r)	15 M W e	Th+ ²³⁵ U Driver Fuel, Coated fuel particles Oxide & dicarbides	1967 - 1988
THTR, Germany	H T G R Power (Pebble Type)	300 M W e	T h + ²³⁵ U, D river Fuel, C oated fuel particles O xide & dicarbides	1985 - 1989
Lingen, Germany	BWR Irradiation -	60 M W e	T est F u el (T h , P u) O $_2$ p elle ts	Terminated in 1973
Dragon, UK OECD-Euratom also Sweden, Norway & Switzerland	testing H T G R E xperim ental (Pin-in-B lock D esign)	20 M W th	T h + ²³⁵ U D river Fuel, C oated fuel particles D icarbides	1966 - 1973
Peach Bottom, USA	H T G R E x perim ental (Prism atic	40 M W e	T h + ²³⁵ U D river Fuel, C oated fuel particles O xide & D icarbides	1966 - 1972
Fort St V rain, U S A	H T G R Power (Prismatic Block)	330 M W e	T h + ^{2 3 5} U D river Fuel, C o a ted fuel particles D ic arbide	1976 - 1989
M S R E O R N L , U S A	MSBR	7.5 M W th	^{2 3 3} U Molten Fluorides	1964 - 1969
Borax IV & Elk River Reactors, USA	BWRs (Pin Assemblies)	2.4 M W e 24 M W e	Th+ ²³⁵ U Driver Fuel Oxide Pellets	1963 - 1968
Shippingport & Indian Point, USA	LWBR PWR (PinAssemblies)	100 M we 285 M W e	T h + ^{2 3 3} U D river Fuel, O x ide Pellets	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$
SUSPOP/KSTR KEMA, Netherlands	A queous H om ogenous Suspension (Pin Assem blies)	1 MWth	Th + HEU Oxide Pellets	1974 - 1977
NRU & NRX, Canada	M T R (Pin Assemblies)		$\begin{array}{c} T & h + {}^{2} {}^{3} {}^{5} U \\ T & e {}^{s} t & F {}^{u} e {}^{l} \end{array}$	Irradiation – testing of few
KAMINI,	M T R T h e r m a l	30 kW th 40 MW t	Al- ²³³ U Driver Fuel 'J' rod of Th & ThO ₂	A ll three research reactors
CIRUS, &		100 M W t	'J' rod of ThO ₂	in operation
D H R II V A India K A P S 1 & 2 , K G S 1 & 2 , R A P S 2 ,3 & 4 , India	PHWR (PinAssemblies)	220 M W e	ThO ₂ Pellets For neutron flux flattening of initial core after start-up	Continuing in all new PHWRs
FBTR, India	LMFBR (PinAssemblies)	40 M W t	ThO ₂ blanket	In operation





Technical document on Thorium fuel cycle options: Potential benefits and challenges

- RATIONALE FOR THORIUM-BASED FUEL CYCLES
- IMPLEMENTATION SCENARIOS AND OPTIONS
- CURRENT INFORMATION BASE
- FRONT-END ISSUES AND CHALLENGES
- BACK-END ISSUES AND CHALLENGES
- PROLIFERATION RESISTANCE
- ECONOMIC ISSUES



IMF Goal and Objective

- Desired goal: eliminate plutonium excesses and minor actinides
- Desired objective: use them to produce energy in reactors





Examples of Inert Matrix

Inert Matrix type	Inert Matrix formula
Element	C, Mg, Al, Si, Cr,V, Zr, Mo, W
Inter-metallics	AlSi, AlZr, ZrSi
Alloy	Stainless steel, zirconium alloys
Carbide	SiC, TiC, ZrC
Nitrides	AIN, TiN, ZrN, CeN,
Binary oxide	MgO, Y ₂ O ₃ , ZrO ₂ , CeO ₂
Ternary oxide	MgAl ₂ O ₄ , Y ₃ Al ₅ O ₁₂ , ZrSiO ₄
Oxide solid solution	$Y_{y}Zr_{1-y}O_{2-y/2}, Mg_{(1-x)}AI_{(2+x)}O_{(4-x)}$

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Examples of Inert Matrix design and additives

Design

Solid solution Cercer

Cermet

Metmet

Composition

 $An_{z}Y_{y}Pu_{x}Zr_{1-y}O_{2-\psi}^{*}$

 $MgAl_2O_4 - Y_yPu_xZr_{1-y}O_{2-y/2}^*$

 $Zr - Y_y Pu_x Zr_{1-y-x} O_{2-y/2}^*$

PuAl₄*-Al





Technical Document on

Viability of Using Inert Matrix Fuel in Reducing Plutonium in Reactors

1. Overview:

2. Potential IMF application; environmental, fuel cycle aspects, economic, nonproliferation aspects, historical before 80's, current programs

- 3. R&D activities for IMF qualification
- 4. Country Specific programs
- 5. International programs: Irradiations
- 6. Overview of IMF workshops
- 7. Outlook and Results



Technical meeting on Current status and future prospects of Gas-Cooled Reactor fuels (Jun 2004)

There were 27 technical presentations from 37 Participants from 17 Member States, EC and 3 IAEA Experts

Technical programme: 1.) Countries Overview 2.) Coated Particle Fuel modelling 3.) Coated Particle Fuel Performance and technology and 4.) Novel ideas and application related to Coated Particle Fuel

Panel Discussion: 1.) Requirements regarding Coated Particle Fuel characteristics for Hydrogen Generation 2.) Desirability of creating a central data-book for coated particle fuel data by the Agency and 3.) Measures for improving international cooperation in Coated Particle Fuel development





Adaptability of MA-fuel fabrication to remote handling in shielded cells

Oxide	Powder metallurgy of pellet fuel	Not easily adaptable to remote handling	SOL-GEL coprecipitation of TRUs from nitrate solution Alternative fuel forms such as Vibro-pac or sphere-pac
Nitride	Powder metallurgy by nitration + carbothermic reduction	Need for very high temperature and very high affinity of Zr for oxygen	Use of SOL-GEL with direct compact could reduce number of steps
Metal-alloy	Injection- casting of alloy fuels	Americium vaporization	Americium trapping system for recover and reuse. Alternatively powder metallurgical method viz., mixing, pellet making & sintering



MA-Fuel development research

- MA-fuel fabrication processes: Powder-metallurgy, Injection casting, SOLGEL, Carbothermic reduction, Vibro-pac, Coated particle development, direct coagulation casting, etc.,
- MA-fuels (targets) and fuel types & forms- catalogue (CERMET, CERCER, IMF, & METMET, as well as MA-MOX, Coated particle, Metal-alloys & Nitride)
- Potential innovative methods for remote fuel fabrication with reduced losses; Waste-generation at the fuel fabrication stages
 - Disposition / flexibility to reprocessing /multi-recycle
 - Advanced core materials
 - Pertinent thermo-physical properties for fuel design, fabrication process and performance Irradiation screening tests, modeling, fuel design and post-irradiation examination

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Minor Actinide Property Database (MADB) Under Preparation

- Bibliographic database on thermodynamic and thermophysical properties of minor actinide (Np, Am, Cm) metals, alloys and compounds
- Access on the internet with some search and filter capabilities (www-nfcis.iaea.org)

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International Atomic End	rgy Agency		NFCIS UDEPO			
MADB Minor	MADB Minor Actinide Property Database					
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Alloys	Alloy Al-Cm	<u>Other</u>	<u>Dissolution</u>			
Alloys	<u>Ally Ni-Cm</u>	<u>Other</u>	<u>Dissolution</u>			
Alloys	<u>Am₁₁Cd₄₅</u>	Lattice Parameter	Lattice parameter			
Alloys	<u>Am-Cd</u>	<u>Crystal Structure</u>	Structure			
Alloys	AmCd ₃	Lattice Parameter	Lattice parameter			
Alloys	AmCd ₆	Lattice Parameter	Lattice parameter			
Alloys	<u>Np₂Ni₁₇</u>	<u>Crystal Structure</u>	Structure			
Alloys	<u>Np₂Ni₁₇</u>	Lattice Parameter	Lattice parameter			

Relation between Process Losses in Nuclear Fuel cycle and Environmental Impact







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Target Values for An-Losses to Achieve a Transmutation Rate of about 100



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Review of pyro-chemical methods

	Metal Electro- refining	Oxide Electro- winning	Chloride / fluoride volatility	Nitride electro- refining	Pyro for HLLW	Novel and auxiliary methods
1. Fuel form	Zr-based actinide alloys	Mixed actinide oxides	ADS targets & TRISO fuel	Actinide nitride	Link from aqueous methods	Plasma process Li reduction
2. MA loading	Fuel fabrication	Different steps	Ideal for MAs processing	High MA loading		
3. Scale	Semi-plant	Semi plant	Laboratory	Laboratory	Laboratory	Laboratory
4. Some of the interested Member States	USA, Japan, EU, Korea, India	Russia, Japan,EU	USA, EU	Japan	USA, Russia, Japan, EU	USA, Russia, Japan, EU



Review of other aqueous partitioning methods

- DIAMEX process to extract Am+Cm
- **SESAME process for Am/Cm separation**
- NEXT process includes uranium crystallization step
- •
- GANEX Process (Actinide Group Separation method)
- UREX process



Combination of Pyro-chemical and Wet process

	Pyro-chemical process	Wet process
Product Purity	Mixture of TRU - Low	High Single elements (U,Pu,Np,Am,Cm)
Process	Batch	Continuous
Throughput	Low	High
Process losses	To be established	Established

Steps to achieve synergetic combination of both processes

Coordinated Research Project (CRP)

"Study of Process Losses in Separation Processes in Partitioning and Transmutation (P&T) Systems in view of Minimizing Long Term Environmental Impacts"

- 1st Scope Definition Meeting Oct. 2002, Vienna
- China, Czech Republic, Germany, India, Japan, Republic of Korea, Russian Federation and USA
- 1st Research Coordination Research Meeting -Dec 2003, Vienna
- 2nd Research Coordination Research Meeting Dec 2004, Czech Republic





CRP Objectives

- Minimization of losses
- Advanced characterization methods for actinides
- Separation criteria to minimize environmental impact
- List of critical radio-nuclides
- Defining environmental impact associated with partitioning and transmutation processes
- Defining proliferation resistance attributes



Minimization of losses

Studies on minimization of process losses in separation processes define the scope in reducing actinides losses in the waste fraction and in-turn minimizes the ultimate environmental impact. For any Partitioning process the acceptable losses to the waste determine the separation criteria. To achieve public acceptance for nuclear energy generation with transmutation, the reduction of long living radiotoxicity must be impressive.

- a) Identify Sources of losses of long living radiotoxic waste
- b) Understand chemical forms of actinides and others in process
- c) Assess the behavior of actinides and FP in the process, such as volatilization, precipitation, etc.,



Advanced characterization methods for actinides

Advanced characterization methods should be developed for transuranic elements to support the minimization of environmental impact. Procedures for measuring the possible material hold-up and reduction of radio-toxicity with accuracy should be inventorized and improved and analytical techniques should be developed

With the following objectives

- 1. Environmental release (discard of long living radiotoxic nuclides)
- 2. fissile material safeguards (flow of fissile material across the plant)
- 3. criticality control (tracking of fissile material masses in the different criticality zones)

Characterization methods for

- Hold up of nuclide masses in process units
- Characterisation of discards in its different forms namely i) remains in hulls ii) fines from shearing iii) dross in crucibles



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Separation criteria to minimize environmental impact

Establishment of criteria for the separations processes in order to realize the target environmental benefits. Conflict between ecological and economic impact by P&T. Cost increases exponentially with the reduction of losses to the waste.

Criteria for Separation

- Establishment of quantitative relationship between environmental impact and separation criteria
- How high separation efficiency should be required
- Possible method for separation and technological feasibility
- How should be treated or utilized the separated species

Additional Considerations

- Handling facilities for treatment and treated material processing
- Dose considerations
- Reactor fuel qualification
- Reduction of depth of disposal

Reduction of Radiotoxicity





List of critical radio-nuclides

Identify and List the Species

- Species to contribute the intrinsic reduction of toxicity
- Species to affect on the safety assessment with dose rate
- Species to contribute the volume reduction of waste to be disposed at deep geologic formation
- Species for possible utilization





Conclusions

- Identification of sources of loss (e.g. of long living radiotoxic waste)
 - Understanding of chemical processes (e.g. of actinides)
 - Understanding of the physical processes (e.g. actinides and FP taking part in volatilization, precipitation, etc.,)
 - Adaptation of the processes to suppress losses
- Development of advanced characterization methods for P&T;
- Group separation of all Actinides
- Integration of treatment and re-fabrication processes and technologies
- Innovative fuel cycles such as thorium fuel cycle
- Establishing a knowledge database for development of partitioning and transmutation technology matured towards environment friendly fuel cycles.
- Development of Regional and International approach



Thank you for your kind attention





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Agency Coordination in Fuel Cycle Studies

Completing

- Thorium Fuel Cycle: Potential Benefits and Challenge
- Viability of Using IMF in Reducing the Plutonium in Reactors
- Current Status and Future Trends of HEU

Ongoing

- Management of RepU
- Process and property of minor actinide compounds and alloys for nuclear fuel and target for incineration in thermal and fast neutron spectra
- Current status and future prospects of gas cooled reactor fuels
- Minor actinide database for advanced fuel cycles
- Study of process-losses in separation processes in P&T systems in view of minimizing long term environmental impacts

Commencing

• Current status and future perspective of liquid metal-cooled reactor fuel cycle

The GANEX concept : Group ActiNides EXtraction





Reduced Pu and Minor Actinide Inventory with LMFR Fuel Cycle



