The Abdus Salam
International Centre for Theoretical Physics

United Nations
Educational, Scientific
and Cultural Organization

International Atomic
Energy Agency

Workshop on

ROLE OF PARTITIONING AND TRANSMUTATION IN THE MITIGATION OF THE POTENTIAL ENVIRONMENTAL IMPACTS OF NUCLEAR FUEL CYCLE

20 - 24 November 2006

ICTP - Trieste, Italy

1774/13

Conventional and Advance Fuels for Nuclear Power Reactors

C. Ganguly
IAEA Vienna

ICTP Workshop

ROLE OF PARTITIONING AND TRANSMUTATION IN THE MITIGATION OF THE POTENTIAL ENVIRONMENTAL IMPACTS OF NUCLEAR FUEL CYCLE

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Conventional and Advance Fuels for Nuclear Power Reactors

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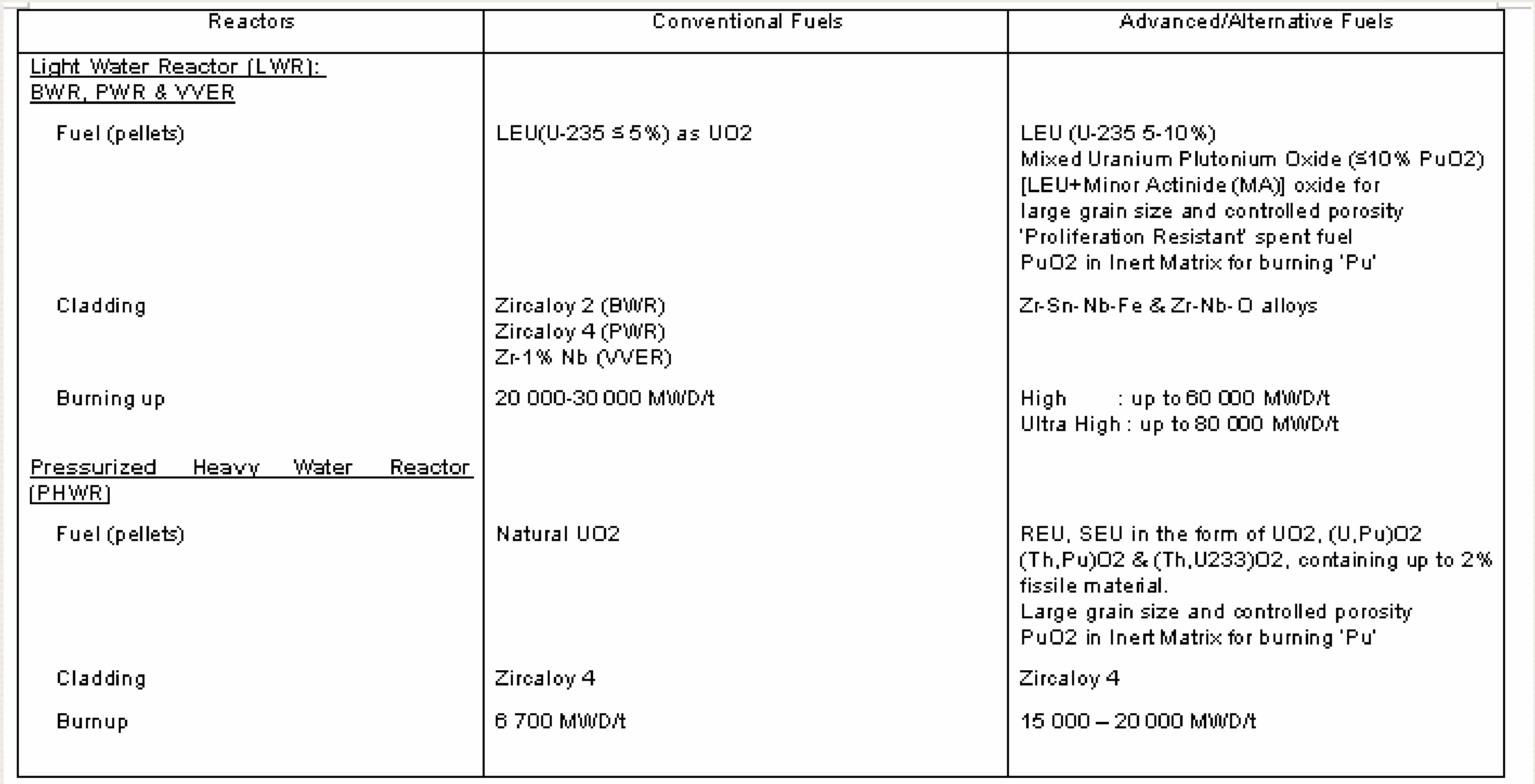
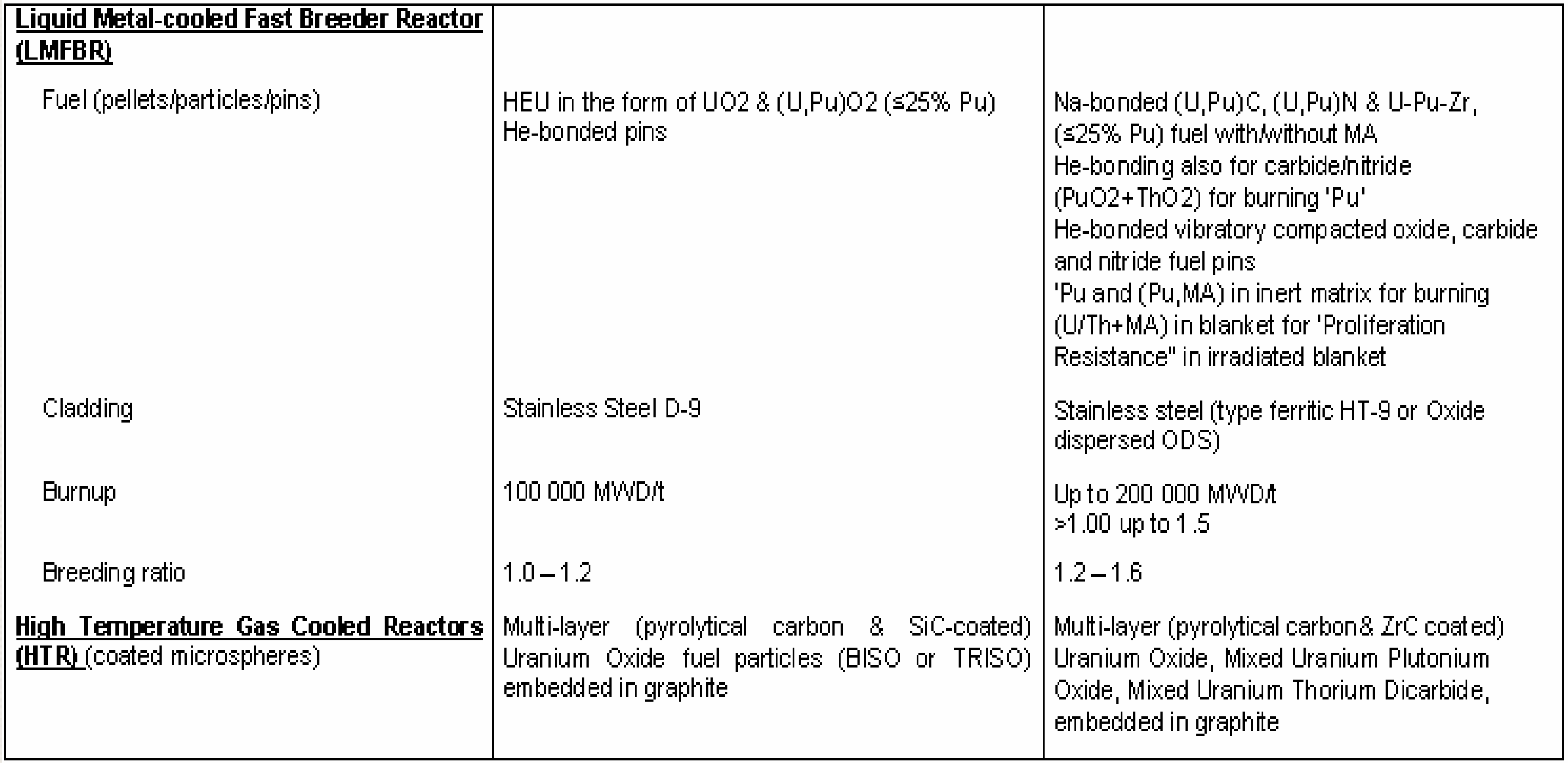
International Atomic Energy Agency

Conventional & Advanced Fuels For Nuclear Power Reactors



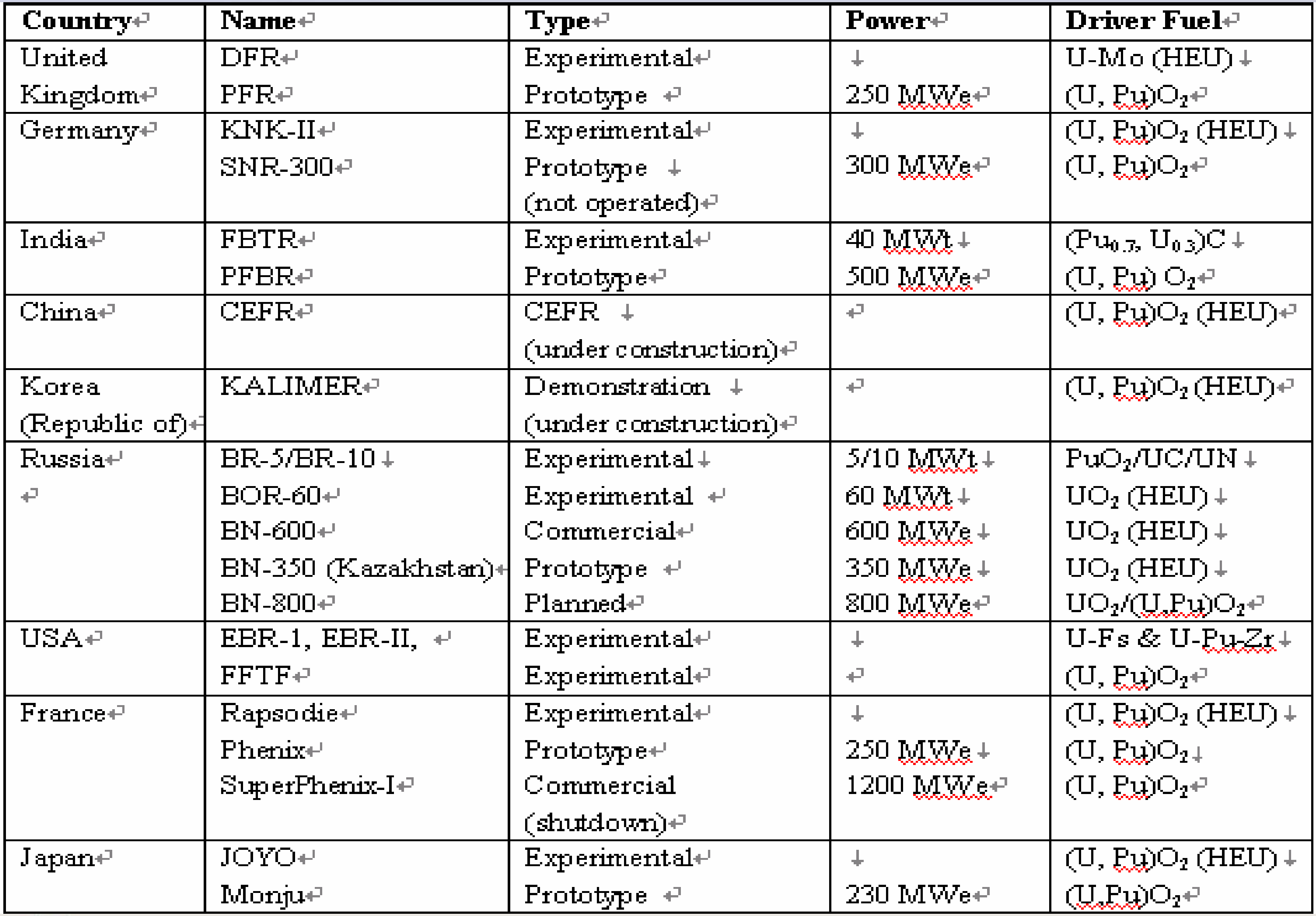
Reactors	Conventional Fuels	Advanced/Alternative Fuels
Light Water Reactor (LWR): BWR, PWR & VVER		
Fuel (pellets)	LEU(U-235 ≤ 5%) as UO2	LEU (U-235 5-10%) Mixed Uranium Plutonium Oxide (≤10% PuO2) [LEU+Minor Actinide (MA)] oxide for large grain size and controlled porosity 'Proliferation Resistant' spent fuel PuO2 in Inert Matrix for burning 'Pu'
Cladding	Zircaloy 2 (BWR) Zircaloy 4 (PWR) Zr-1% Nb (VVER)	Zr-Sn-Nb-Fe & Zr-Nb-O alloys
Burning up	20 000-30 000 MWD/t	High : up to 60 000 MWD/t Ultra High : up to 80 000 MWD/t
Pressurized Heavy Water Reactor (PHWR)		
Fuel (pellets)	Natural UO2	REU, SEU in the form of UO2, (U,Pu)O2 (Th,Pu)O2 & (Th,U233)O2, containing up to 2% fissile material. Large grain size and controlled porosity PuO2 in Inert Matrix for burning 'Pu'
Cladding	Zircaloy 4	Zircaloy 4
Burnup	6 700 MWD/t	15 000 – 20 000 MWD/t

Conventional & Advanced Fuels For Nuclear Power Reactors



Liquid Metal-cooled Fast Breeder Reactor (LMFBR)		
Fuel (pellets/particles/pins)	HEU in the form of UO2 & (U,Pu)O2 (≤25% Pu) He-bonded pins	Na-bonded (U,Pu)C, (U,Pu)N & U-Pu-Zr, (≤25% Pu) fuel with/without MA He-bonding also for carbide/nitride (PuO2+ThO2) for burning 'Pu' He-bonded vibratory compacted oxide, carbide and nitride fuel pins 'Pu and (Pu,MA) in inert matrix for burning (U/Th+MA) in blanket for 'Proliferation Resistance" in irradiated blanket
Cladding	Stainless Steel D-9	Stainless steel (type ferritic HT-9 or Oxide dispersed ODS)
Burnup	100 000 MWD/t	Up to 200 000 MWD/t >1.00 up to 1.5
Breeding ratio	1.0 – 1.2	1.2 – 1.6
High Temperature Gas Cooled Reactors (HTR) (coated microspheres)	Multi-layer (pyrolytical carbon & SiC-coated) Uranium Oxide fuel particles (BISO or TRISO) embedded in graphite	Multi-layer (pyrolytical carbon & ZrC coated) Uranium Oxide, Mixed Uranium Plutonium Oxide, Mixed Uranium Thorium Dicarbide, embedded in graphite

Fast Reactors in the world and their driver fuels



Country	Name	Type	Power	Driver Fuel
United Kingdom	DFR PFR	Experimental Prototype	 250 MWe	U-Mo (HEU) (U, Pu)O$_2$
Germany	KNK-II SNR-300	Experimental Prototype (not operated)	 300 MWe	(U, Pu)O$_2$ (HEU) (U, Pu)O$_2$
India	FBTR PFBR	Experimental Prototype	40 MWt 500 MWe	(Pu$_{0.7}$, U$_{0.3}$)C (U, Pu) O$_2$
China	CEFR	CEFR (under construction)		(U, Pu)O$_2$ (HEU)
Korea (Republic of)	KALIMER	Demonstration (under construction)		(U, Pu)O$_2$ (HEU)
Russia	BR-5/BR-10 BOR-60 BN-600 BN-350 (Kazakhstan) BN-800	Experimental Experimental Commercial Prototype Planned	5/10 MWt 60 MWt 600 MWe 350 MWe 800 MWe	PuO$_2$/UC/UN UO$_2$ (HEU) UO$_2$ (HEU) UO$_2$ (HEU) UO$_2$/(U,Pu)O$_2$
USA	EBR-1, EBR-II, FFTF	Experimental Experimental		U-Fs & U-Pu-Zr (U, Pu)O$_2$
France	Rapsodie Phenix SuperPhenix-I	Experimental Prototype Commercial (shutdown)	 250 MWe 1200 MWe	(U, Pu)O$_2$ (HEU) (U, Pu)O$_2$ (U, Pu)O$_2$
Japan	JOYO Monju	Experimental Prototype	 230 MWe	(U, Pu)O$_2$ (HEU) (U,Pu)O$_2$

FUTURE CHALLENGES – FUEL MATERIALS

Properties	$(U_{0.8}Pu_{0.2})O_2$	$(U_{0.8}Pu_{0.2})C$	$(U_{0.8}Pu_{0.2})N$	U-19Pu-10Zr
Theoretical Density g/cc	11.04	13.58	14.32	15.73
Melting point ºK	3083	2750	3070	1400
Thermal conductivity (W/m ºK) 1000 K 2000 K	2.6 2.4	18.8 21.2	15.8 20.1	40
Crystal structure	Fluorite	NaCl	NaCl	γ
Breeding ratio	1.1 - 1.15	1.2 – 1.25	1.2 - 1.25	1.35 - 1.4
Swelling	Moderate	High	High (?)	High
Handling	Easy	pyrophoric	Inert atmos	Inert atmos
Compatibility - clad coolant	average average	Carburisation good	good good	eutectics good
Dissolution & reprocessing amenability	Good	Demonstrated	risk of C^{14}	Pyro-reprocessing
Fabrication/Irradiation experience	Large Good	limited	very little	limited

Innovative Back-end Technology of Fuel Cycle – Developments in Japan

Reprocessing	Simplified PUREX Process	Alternative Aqueous Process	Oxide – Electrowinning Process	Metal – Electrorefining Process
	Dissolution	Direct Extraction	Uranium Electrowinning	Oxide Reduction
U Recovery	Crystallization			U Electrorefining (solid cathode)
U, Pu,MA Recovery	Single Cycle Extraction	Supercritical Fluid Extraction $TBP-CO_2-HNO_3$	MOX Electro-codeposition	U, Pu, MA Electrorefining (Liq. Cd cathode)
MA Recovery	SETFICS Process /TRUEX Process	Ion Exchange / Amine Extraction	MA Electrowinning	Cd Extraction (pyro contactor)

Oxidation

Fuel Refabrication	Pellet short Process	Sphere Packing Process	Vibro -Packing Process	Casting Process
	Simplified Pelletizing	Gelation (MOX– MA)	Granulation (MOX,MN+MA)	Casting (U,Pu -Zr-MA)
	Stacking	Vibration Packing	Vibration Packing	Stacking
	MOX Fuel		MOX Fuel	Metal Fuel

🟦 :MOX Aqueous Recycle :MO🟨 –Aqueous Recycle :Metal Non🟪 Aqueous Recycle

SUMMARY OF IAEA CONSULTANCY MEETING (Dec. 2004) ON LIQUID METAL-COOLED FAST REACTOR (LMFR) FUEL CYCLE

Russian Federation

PYROCHEMICAL REPROCESSING

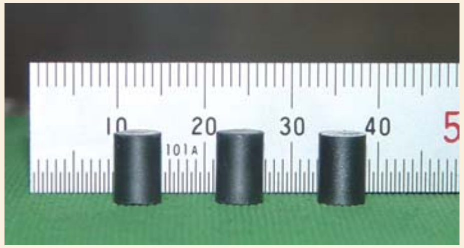
Basic research of the molten salt systems allowed for the development of technological processes for production of granulated uranium and plutonium oxides and mixed uranium and plutonium oxides. A distinctive feature of the pyrochemical technology is a possibility to perform all deposit production operations in one facility – a chlorinator-electrolyzer

PROCESS STEPS

- ***Dissolution of initial products or spent nuclear fuel in molten salts***
- ***Recovery of crystal plutonium dioxide or electrolytic plutonium and uranium dioxides from the melt***
- ***Processing of the cathode deposit and production of granulated fuel***

CANDIDATES OF FUEL CYCLE SYSTEMS

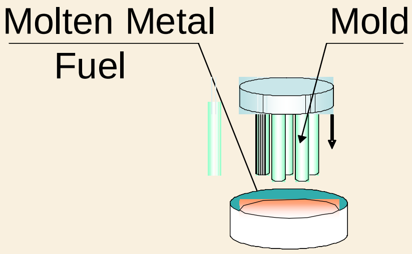
Fuel Fabrication



Simplified Pelletizing

[Special Features]

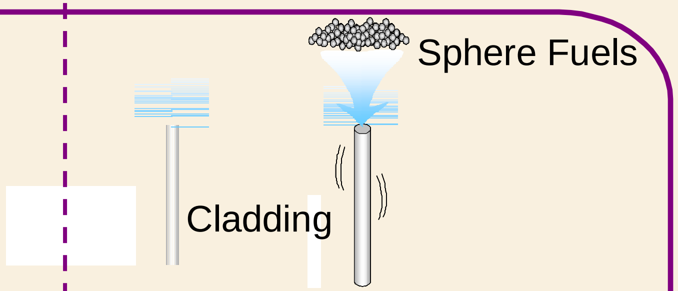
- **Abundant Experiences**
- **Rationalize to 1/3 Processes**



Injection Casting

[Special Features]

- **Simple Process**
- **Small Fabrication Equipment**

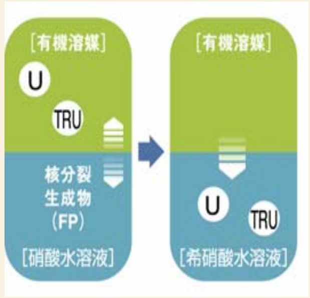


Sphere and Vibro-Packing

[Special Features]

- **Simple Process**
- **Possibility of High Economics**

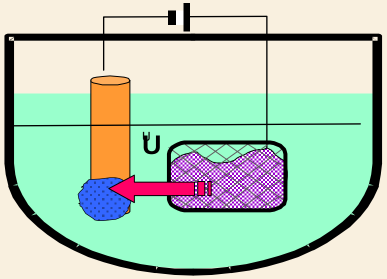
Reprocessing



Advanced Aqueous

[Special Features]

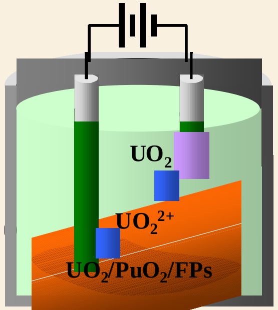
- **Abundant Experiences**
- **Higher Technical Realization**



Metal Electro-refining

[Special Features]

- **Possibility of Higher Economics**
- **Accomplishments in the U.S.**



Oxide Electro-winning

[Special Features]

- **Many Fundamental Subjects**
- **Accomplishments in Russia**

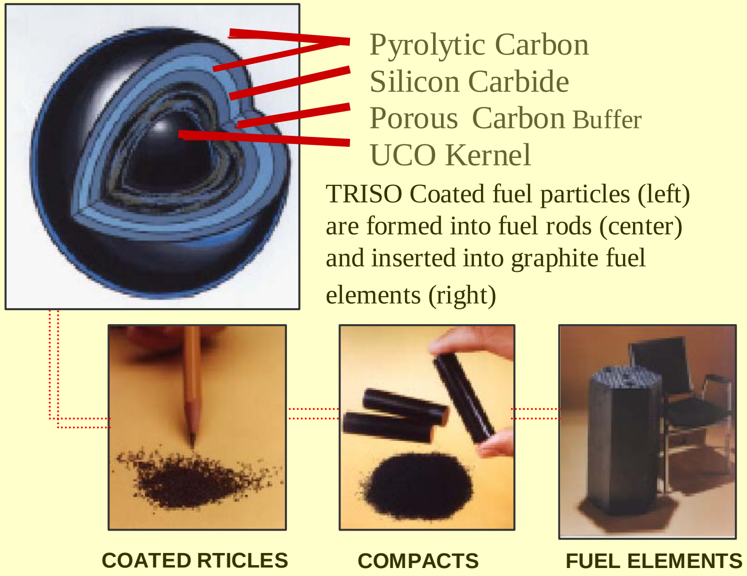
FUEL FORMS

B: Coated Fuel Particles

1. Coated fuel particles for HTGR

Prismatic block

US, Japan, Russia and France



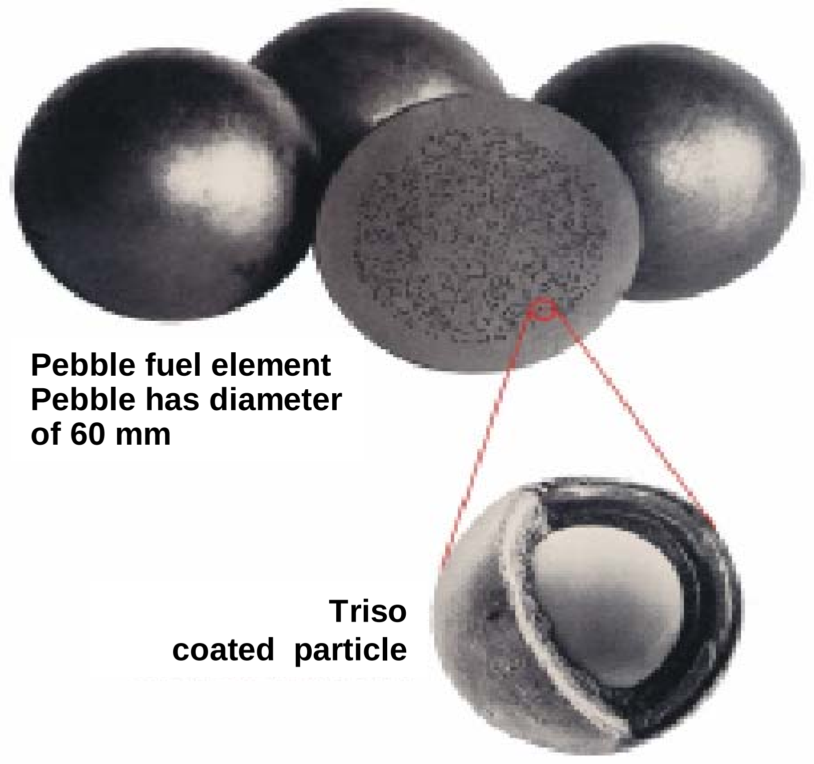
TRISO Coated fuel particles (left) are formed into fuel rods (center) and inserted into graphite fuel elements (right)

2. Fuel particles (dry or wet route) for vibratory compacted fuel pins

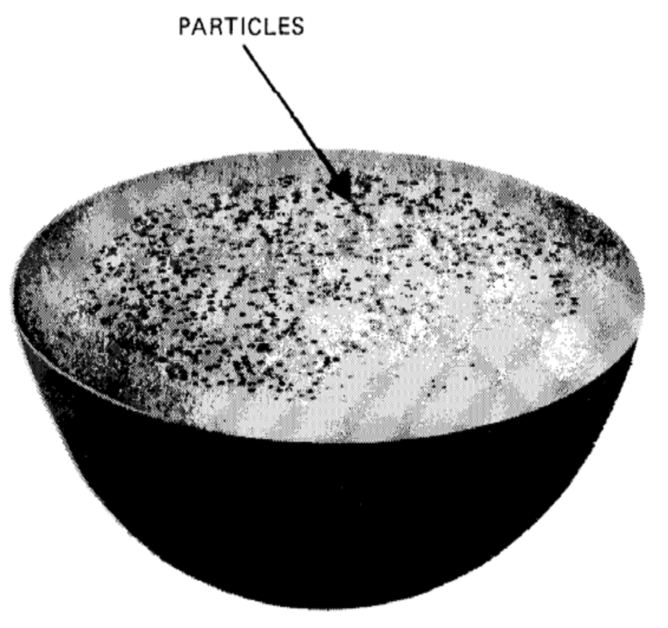
Pebble Bed

coated particle fuels embedded in spherical shape

Germany, South Africa, China

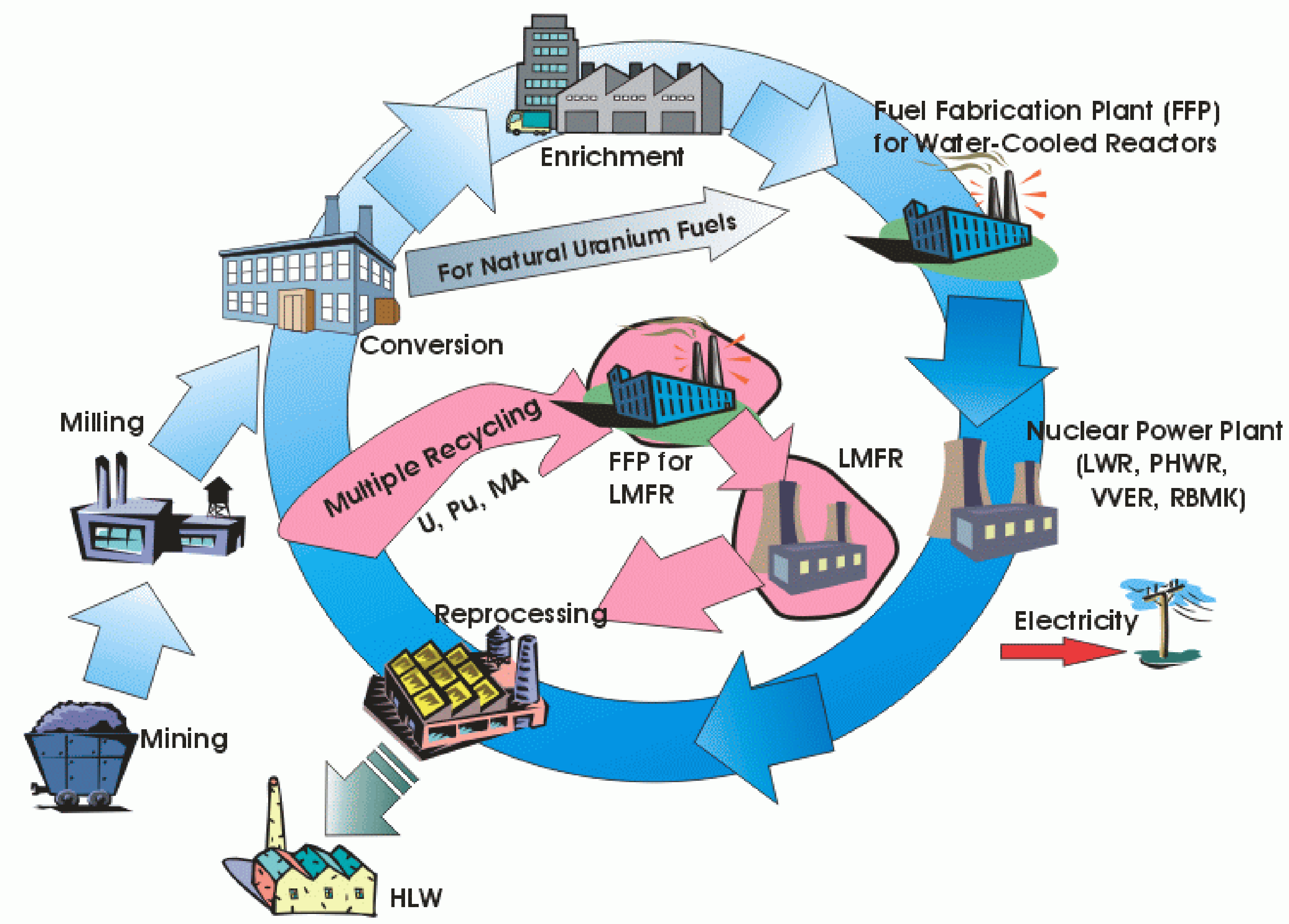


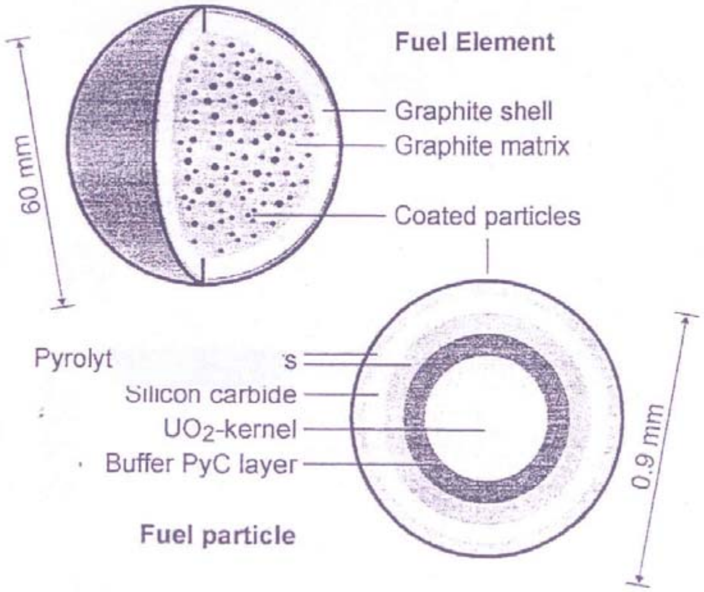
In helium cooled “pebble bed reactors”, fuel particles are contained in graphite pebbles (d= 6 cm)



- ~ 15000 coated fuel particles / pebble
- ~ 800,000 pebbles / core (~ 1/3 without fuel) for a 165 MWe reactor
- on-line re-fueling

Liquid Metal-cooled Fast Reactor Fuel Cycle with multiple recycling of U, Pu and Minor Actinides





COATED FUEL PARTICLES

FUEL PELLETS

COMMERCIAL SPENT URANIUM OXIDE FUEL REPROCESSING PLANTS IN OPERATION AND UNDER CONSTRUCTION IN THE WORLD

Country / Company	Facility / Location	Fuel Type	Capacity (tHM/year)
France, COGEMA	UP2 and UP3, La Hague	LWR	1700
UK, BNFL	Thorp, Sellafield	LWR, AGR	1200
UK, BNFL	B205 Magnox	Magnox GCR	1500
Russian Federation, Minatom	RT-1 / Tcheliabinsk-65 Mayak 400	VVER	400
Japan, JNC	Tokai-Mura	LWR, ATR	90
Japan, JNFL	Rokkasho-Mura (under construction)	LWR	800
India, BARC	PREFRE-1, Tarapur PREFRE-2, Kalpakkam	PHWR PHWR	100 100
China, CNNC	Diowopu (Ganzu)	LWR	25-50

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MIXED URANIUM PLUTONIUM OXIDE (MOX) FUEL FABRICATION FACILITIES

Country / Company	Facility / Location	Fuel Type	Capacity (tHM/year)
France, COGEMA	Cadarache	LWR, FBR	40
France, COGEMA	Marcoule-Melox	LWR	100
Belgium, Belgonucleaire	Dessel	LWR	40
UK, BNFL	Sellafield SMP	LWR	120
UK	Sellafied MDF	LWR	8
Russian Federation, Minatom	Chelyabinsk	FBR	60
Japan, JNC	Tokai-Mura	ATR	10
Japan, JNFL	Rokkasho	LWR	130
India, AFFF, BARC	Tarapur	LWR, PHWR & FBR	

R&D on Advanced LMFR Fuels and Advanced Methods of Fuel Fabrication

- **Ceramic Nuclear Fuels**
  Conventional: (U, Pu)O_2
  Advanced: (U, Pu)C & (U, Pu)N with / without Minor Actinides

- **Advanced methods of fabrication of ceramic fuels:**
  Dust-free advanced fabrication processes like vibratory compaction, vibro-sol & sol-gel microsphere pelletization

- **Metallic Fuels:**
  U-Pu-Zr, Th-U-Pu-Zr & U-Pu (for high breeding)

- **Fuel Cladding, Hexcans & Other Fuel Assembly components:**
  Ferritic stainless steel HT9 & Oxide dispersed stainless steel with minimum radiation damage and void swelling

- **Advanced fabrication processes should be amenable to secured automated fabrication, real-time accounting of special nuclear material and proliferation resistance**

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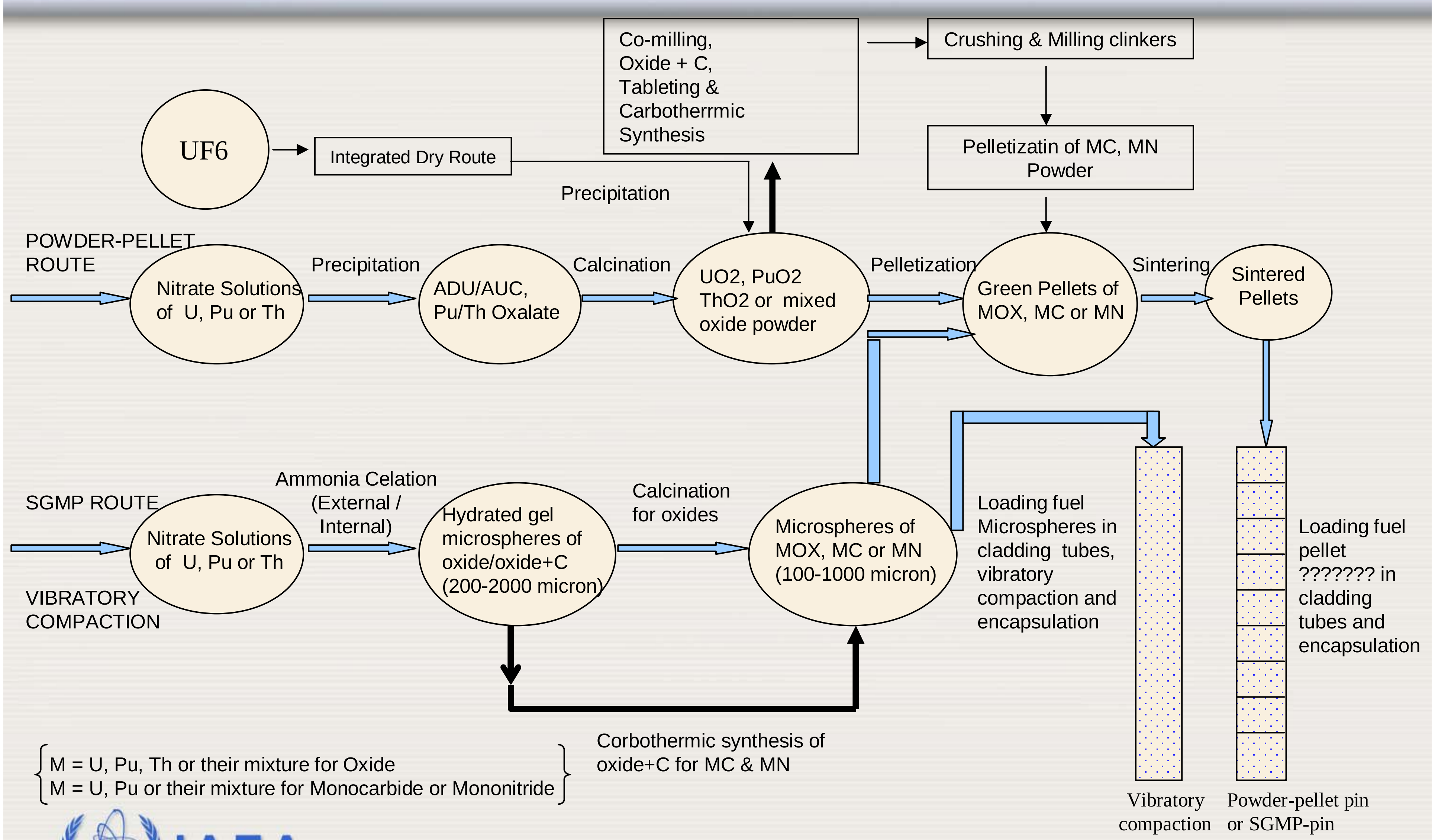
Objectives of advanced methods of fabrication of ceramic nuclear fuel pellets

Safety	Economics	Performance
• Avoid generation and handling of powder of fuels for minimising : - radiotoxic dust hazard - fire hazard (for carbide & nitride fuels) • Fabrication flow sheet should be amenable to automation & remotisation - for minimising personnel exposure to radiation	• Minimise process steps • Reduce fuel synthesis & sintering temperatures • Reduce gas cost during synthesis and sintering - gas purification and recirculation - alternative less expensive gas • Reduce process losses and rejects	• Tailor make fuel microstructure for higher burn up - High density (≥96% T.D.), closed "porosity" and large (>25µ) grain size for LWR & PHWR - Low density (<85% T.D.) "open" porosity and small (<5µ) grain size for LMFBR - Excellent micro-homogenity of fissile material in fuel - avoid fine pores (<1µ) for minimising in-pile densification



Co-milling,
Oxide + C,
Tableting &
Carbotherrmic
Synthesis

Crushing & Milling clinkers

UF6

Integrated Dry Route

Pelletizatin of MC, MN Powder

Precipitation

POWDER-PELLET ROUTE

Nitrate Solutions of U, Pu or Th

Precipitation

ADU/AUC, Pu/Th Oxalate

Calcination

UO2, PuO2 ThO2 or mixed oxide powder

Pelletization

Green Pellets of MOX, MC or MN

Sintering

Sintered Pellets

SGMP ROUTE

Nitrate Solutions of U, Pu or Th

Ammonia Celation (External / Internal)

Hydrated gel microspheres of oxide/oxide+C (200-2000 micron)

Calcination for oxides

Microspheres of MOX, MC or MN (100-1000 micron)

Loading fuel Microspheres in cladding tubes, vibratory compaction and encapsulation

Loading fuel pellet ??????? in cladding tubes and encapsulation

VIBRATORY COMPACTION

Corbothermic synthesis of oxide+C for MC & MN

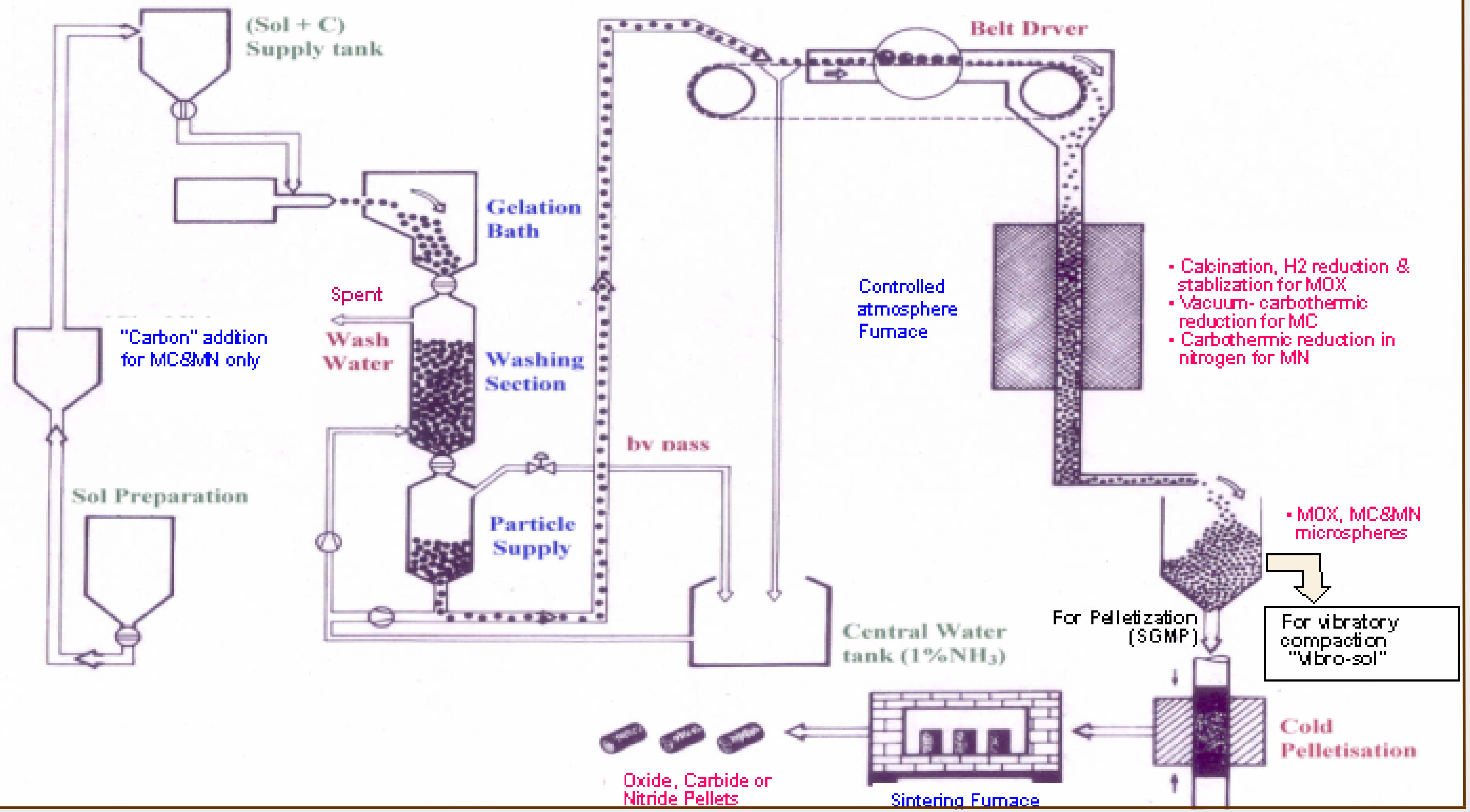
M = U, Pu, Th or their mixture for Oxide
M = U, Pu or their mixture for Monocarbide or Mononitride

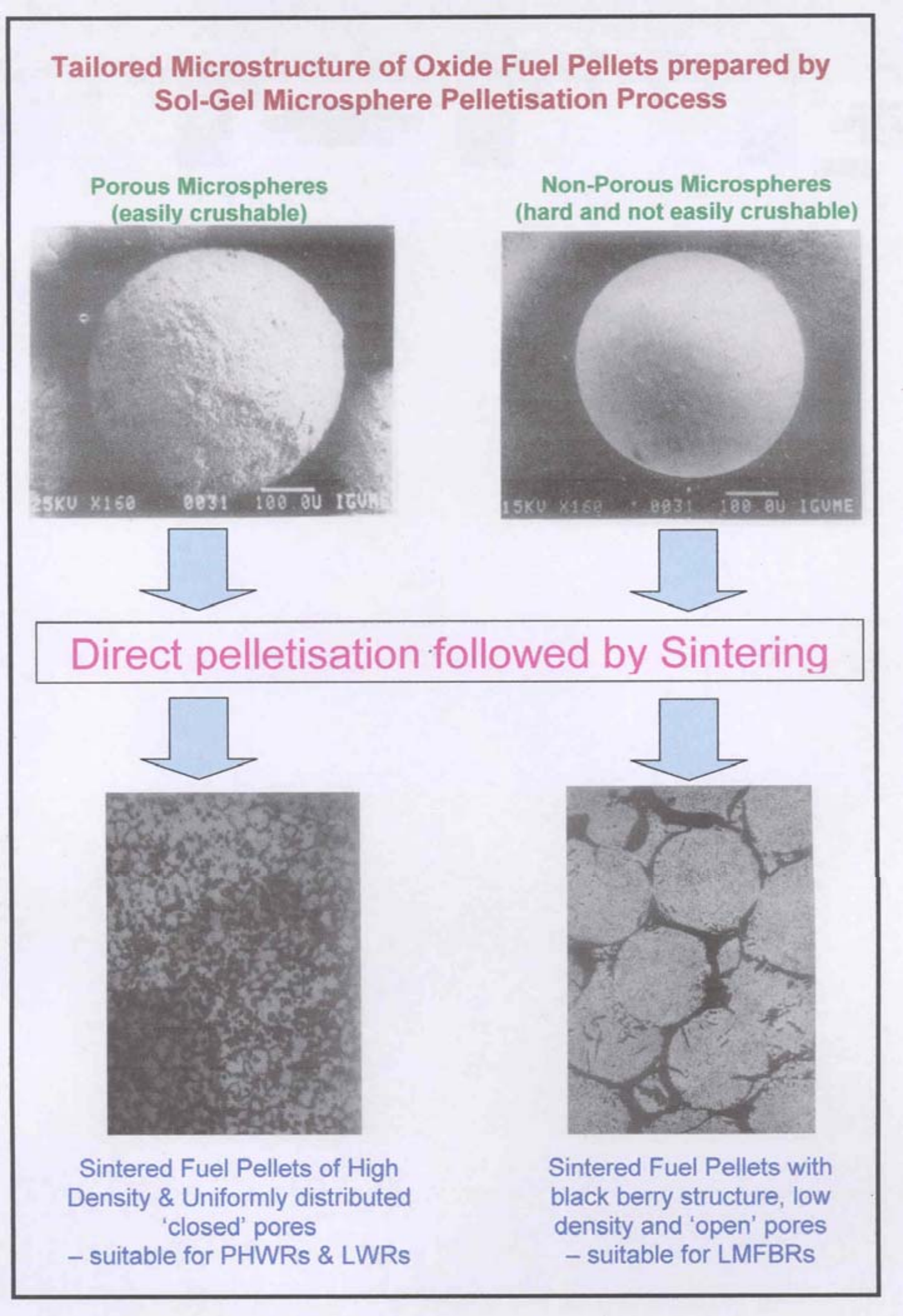
Vibratory compaction

Powder-pellet pin or SGMP-pin

"Sol-Gel-Microsphere Pelletization (SGMP)" and "Vibro-Sol" Processes for Manufacturing Mixed Uranium Plutonium Oxide (MOX), Monocarbide (MC) and Mononitride (MN) Fuels for LMFR

Preparation of Sol-Gel-Microspheres of Oxide or Oxide+C: Ammonia External or Internal Gelation Process





Tailored Microstructure of Oxide Fuel Pellets prepared by Sol-Gel Microsphere Pelletisation Process

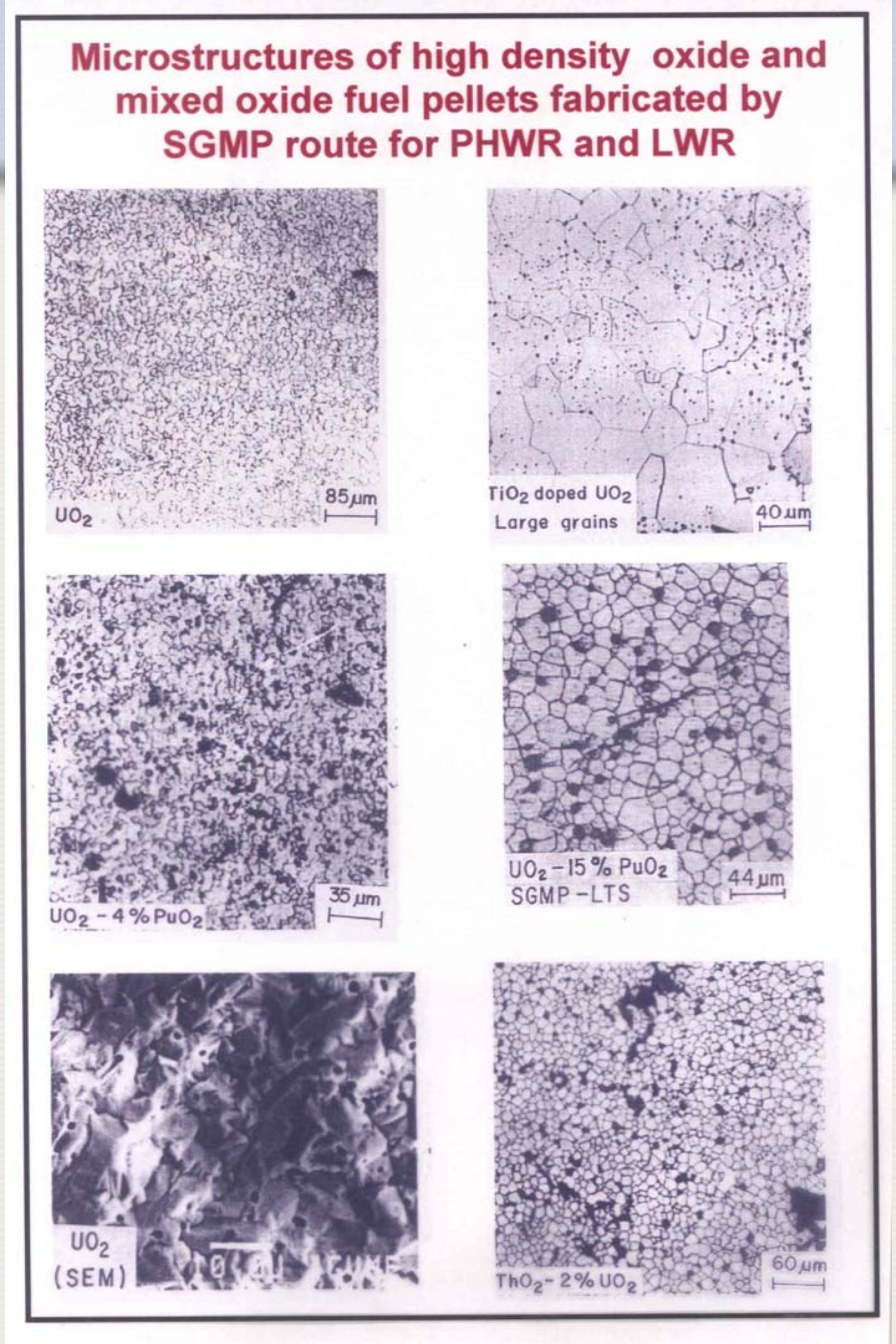
Porous Microspheres (easily crushable)

Non-Porous Microspheres (hard and not easily crushable)

Direct pelletisation followed by Sintering

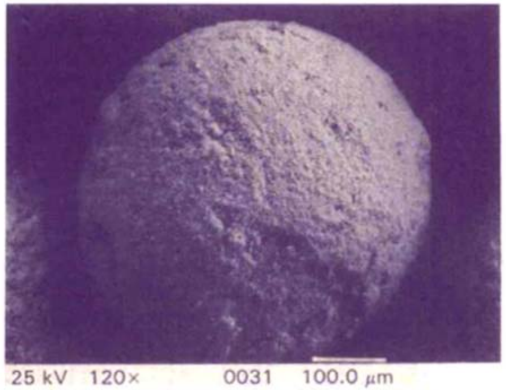
Sintered Fuel Pellets of High Density & Uniformly distributed 'closed' pores – suitable for PHWRs & LWRs

Sintered Fuel Pellets with black berry structure, low density and 'open' pores – suitable for LMFBRs

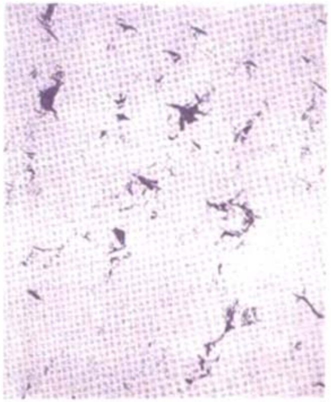


Microstructures of high density oxide and mixed oxide fuel pellets fabricated by SGMP route for PHWR and LWR

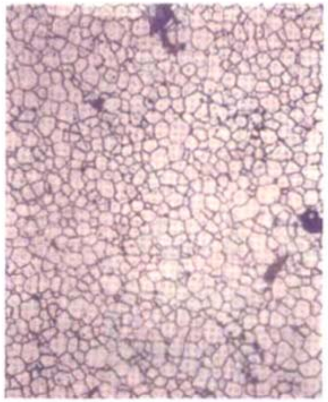
Microstructure & Image Analysis of ThO_2-2%UO_2 prepared by Sol-Gel Microsphere Pelletisation (SGMP) Process



Porous Microsphere (SEM Picture)



As-Polished Microstructure



Etched Microstructure

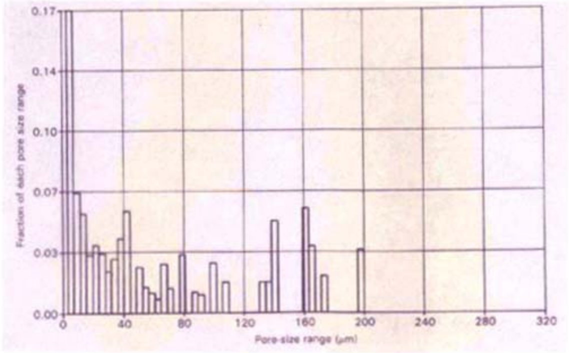


Image Analysis of entire cut section of pellet (for determining undissolved microsphere boundary)

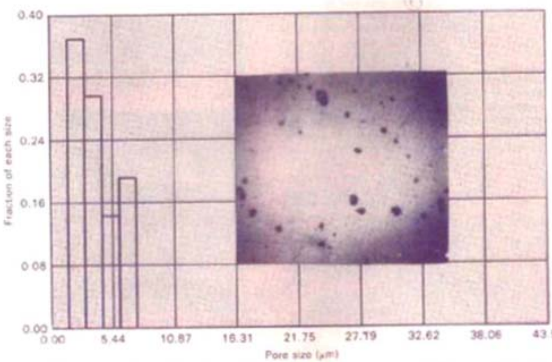


Image Analysis of selected areas (54µ x 54µ) (for average pore size & distribution)

OBJECTIVES OF INERT MATRIX FUEL

- Minimizing “proliferation risk” of plutonium (~ 200 tons of weapon-grade and ~ 1000 tons civilian grade) by using them in nuclear power reactors in operation
- Minimizing “Minor Actinides” (MA: Np, Am & Cm) and in turn radiotoxicity in waste
- In some cases minimizing ‘proliferation risk’ of weapon-grade (> 90 % ^{235}U) uranium (though conventional process is down-blending)

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Inert Matrix

- **Neutron (very low capture and absorption cross-sections)**
- **Chemical compatibility with**
  - → Fuel
  - → Cladding
  - → Coolant
- **Consideration of direct disposal after use**

Fuel

- **'Plutonium form' – alloys and compounds**
- **Utilization of Minor Actinides together with plutonium**
- **Weapon-grade HEU (^{235}U > 90 %) –alloys or compounds**

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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	**AlN, TiN, ZrN, CeN,**
Binary oxide	MgO, Y_2O_3, ZrO_2, CeO_2
Ternary oxide	$MgAl_2O_4$, $Y_3Al_5O_{12}$, $ZrSiO_4$
Oxide solid solution	$Y_yZr_{1-y}O_{2-y/2}$, $Mg_{(1-x)}Al_{(2+x)}O_{(4-x)}$

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Conventional and Advanced Methods of Spent Fuel Reprocessing

AQUEOUS PROCESS :

Dissolution of spent fuel in Nitric acid followed by purification by solvent extraction by adapting the PUREX process, using TriButyl Phosphate (TBP) as solvent, is being used on an industrial scale for reprocessing of spent UO2 and MOX fuels. The PUREX process is not suitable for mixed carbide fuel but could be utilized for reprocessing mixed nitride and metallic fuels.

Modifications are being incorporated in PUREX process to make it proliferation resistant and economic.

PYROPROCESSING :

– **Pyroprocessing involving electrolytic reduction**
This route has been initially developed on a pilot plant scale for reprocessing of spent metallic fuels (U-Zr & U-Pu-Zr) in USA and was successfully extended on a laboratory scale for reprocessing of carbide and nitride fuels. The pyroprocessing route is yet to be adapted on an industrial scale.
In recent years, the Russian Federation has successfully demonstrated the pyroprocessing route for reprocessing of spent oxide fuels on a pilot plant scale.

– **Pryoprocessing involving fluoride volatilization**
The process includes fluorination followed by distillation. The method has so far been demonstrated on a laboratory scale only.

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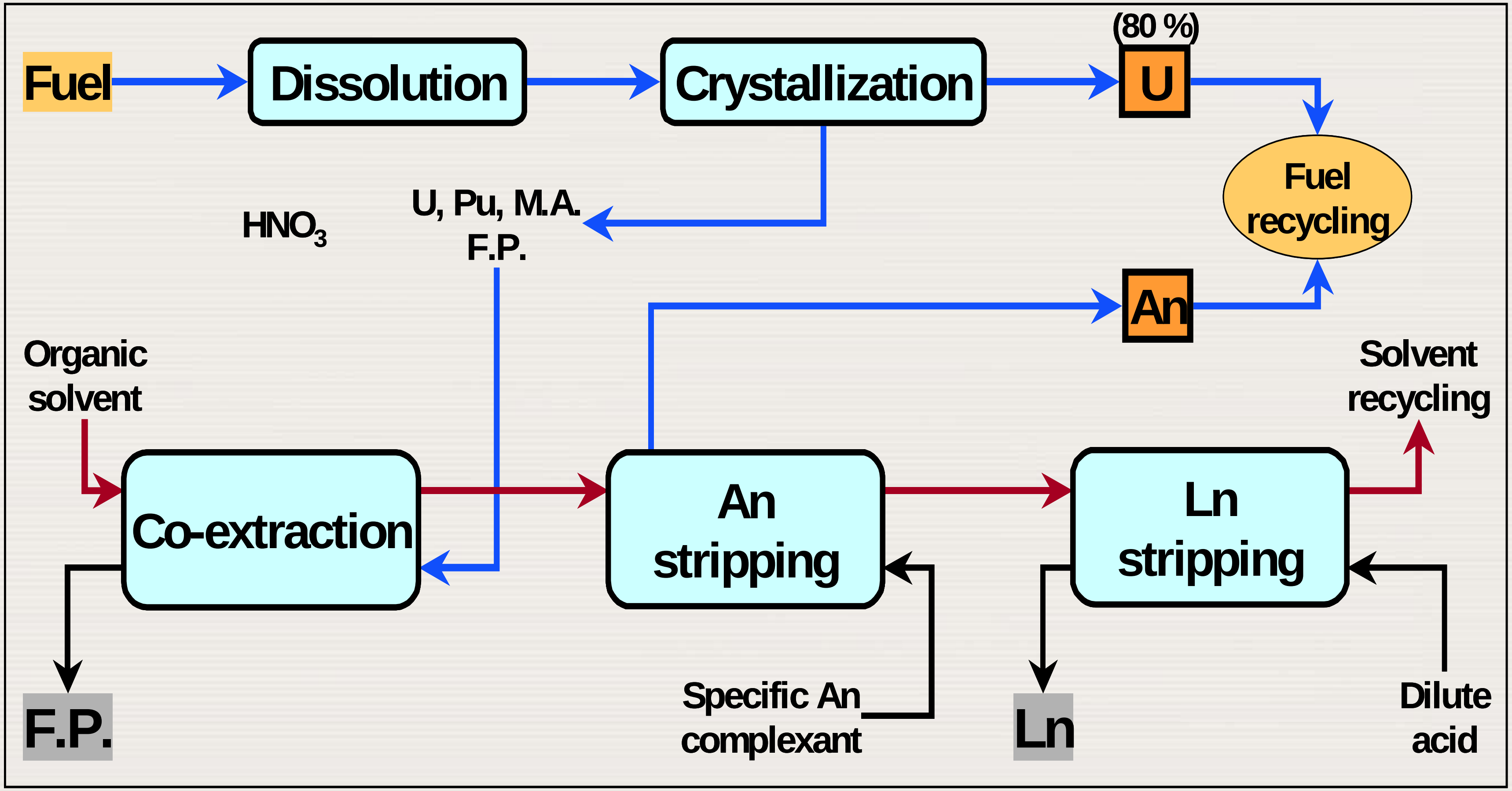
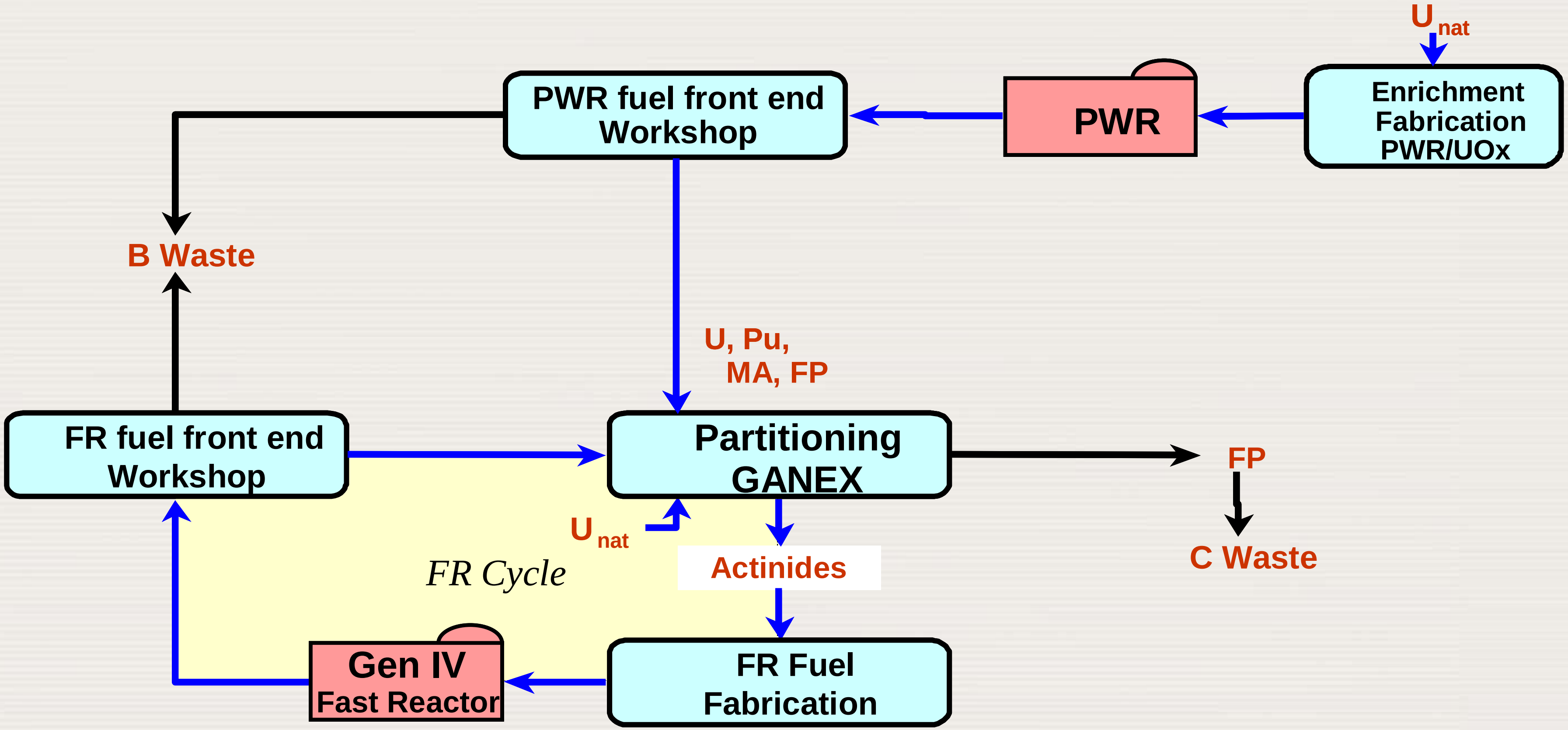
The GANEX concept : Group ActiNides EXtraction

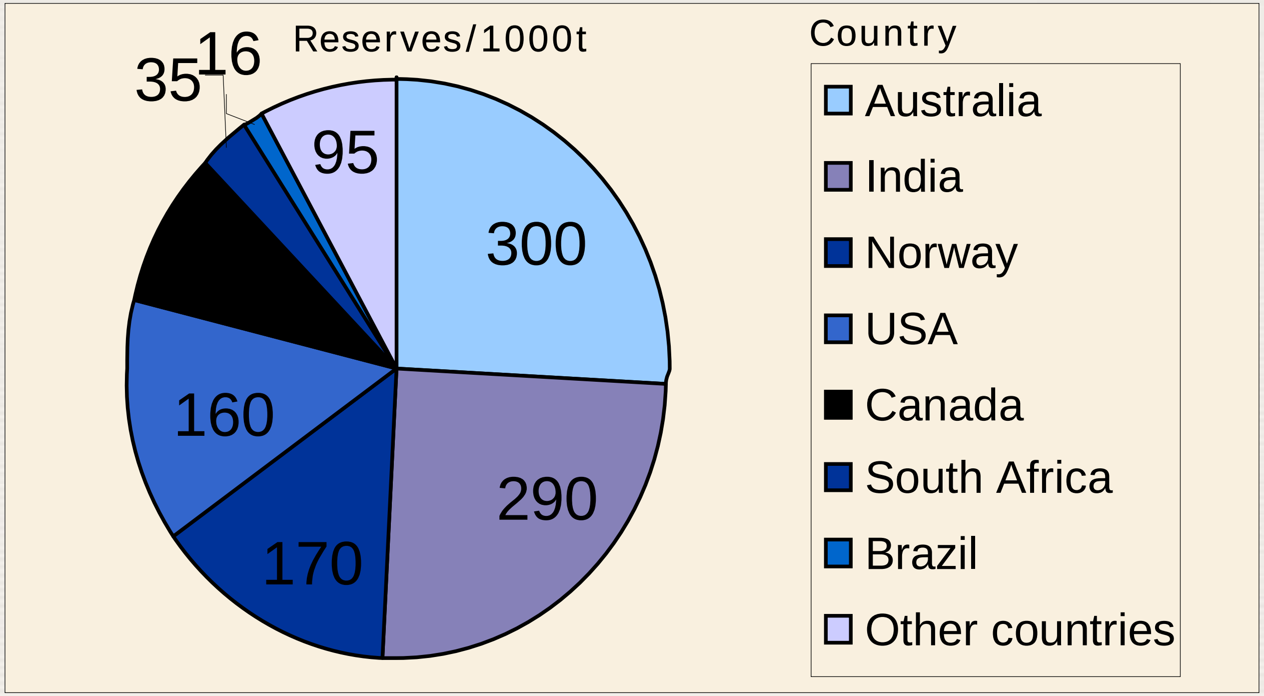


Modular CEA design of a next generation spent fuel treatment

Grouped actinides extraction (GANEX) (M. Delpech et al., CEA)



World Thorium Resources - economically extractable



Country	Reserves / t
Australia	300 000
India	290 000
Norway	170 000
USA	160 000
Canada	100 000
South Africa	35 000
Brazil	16 000
Other Countries	95 000

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