

d Nations ational, Scientific Cultural Organization



Workshop on "Technology and Applications of Accelerator Driven Systems (ADS)"

17 - 28 October 2005

1677/12

P&T as a Waste Management Option

IMPACT of P&T on Nuclear Waste Management

W. Gudowski Royal Institute of Technology, Stockholm, Sweden



P&T as a Waste Management Option

IMPACT of P&T on Nuclear Waste Management

Prof. Waclaw Gudowski KTH, Stockholm wacek@neutron.kth.se





- IAEA
 - Trieste
 - World Nuclear University!!
- Fredric Joliot/Otto Hahn Summer School in Reactor Physics, Fuels & more
- NEA/OECD
- European Activities:
 - ENEN and Eurotrans Trainings
- + more!!!





Program

- Nuclear Fuel Cycle
 - Uranium and Actinides
 - From mine to the final waste disposal
 - Uranium and transuranium elements
 - Uranium Resources
 - Steps in the Nuclear Fuel Cycle
 - Front End of the Fuel Cycle
 - Fuel Utilization
 - Back End of Fuel Cycle
- Reference NFCs
- Quantifying impact
- Examples & Results
- Alternatives





Main problems of the Nuclear Fuel Cycle to be solve today:

• PUBLIC ACCEPTANCE:

- Overcoming public fears, phobias and legitimate concerns
- POLITICS NON PROLIFERATION ISSUES AND CONSTRAINTS
- ECONOMY
- Sustainability & Enivorment
- • • • •
-
- Engineering and Technology





Solving the problems...

....we should focus on THE BIGGEST INDENTIFIED PROBLEMS..however this is not what this lecture is about....



5





MA



- Radioactive metallic element, symbol ⁹²U, actinide. A= 92, at.wt. 238.029.
- Melting temp. 1132° C,
- Boiling temp. 3818° C
- Specific mass 19.05 at 25° C.
- Uranium discovered in 1789 in pitchblende, by the German chemist Martin Heinrich Klaproth.
- Named after the planet Uranus.
- First isolated in the metallic state in 1841.
- The radioactive properties first demonstrated in 1896 by Henri Becquerel.





- **3 crystalline forms,** one formed at about 770° C is malleable and ductile.
- **soluble** in hydrochloric and nitric acids, insoluble in alkalies
- displaces hydrogen from mineral acids and from the salt solutions of such metals as Hg, Ag, Cu, tin, Pt, and Au.
- **burns** in air at 150° to 175° C.
- at 1000° C combines with nitrogen, forms a yellow nitride.
- **oxidation states** of three, four, five, and six.
 - the hexapositive compounds: uranyl trioxide, UO_3 , and uranyl chloride, UO_2CI_2 .
 - Tetrapositive (uranous) : tetrachloride, UCl₄, and uranium dioxide, UO₂. Usually unstable; revert to the hexapositive form when excessively exposed to air.
- Uranyl salts may decompose in the presence of strong light and organic matter





- **U** never occurs naturally in the free state, is found as an oxide or complex salt in minerals: pitchblende and carnotite.
 - Uraninite is the most common primary uranium mineral; others coffinite and brannerite.
- Average concentration in the earth crust about 2.7 ppm.

Average ore grades at operating uranium mines range from 0.03% U to as high as 10% U but are most frequently less than 1% U.

Ranks about **48th** in natural abundance in crustal rocks.

Pure U consists of **99.3** % of the isotope $^{238}U_{,} \sim 0.7$ % of the isotope $^{235}U_{,}$

- Traces of ²³⁴U, formed by radioactive decay of ²³⁸U.
- Isotopes ranging from mass number 222 to 242 are known.

The richest uranium ore (pitchblende), are found chiefly in the Czech Republic, Canada, Congo (Zaire), and the U.S.





Geographical distribution of uranium resources, based on national estimates for selected countries (in million tonnes of uranium).

Region	Reasonably assured	Speculative (potential resources)	Sum
Australia	0.9	3.9	4.8
Former Soviet Union (FSU)	1.8	2.0	3.8
United States	1.7	1.3	3.0
China		1.8	1.8
Mongolia	0.1	1.4	1.5
South Africa	0.3	1.1	1.4
Canada	0.6	0.7	1.3
Sum of listed countries	5.4	12.2	17.6





- Uranium from seawater represents a very large potential additional resource, but at a high price.
- The amounts available are very large. The volume of seawater in the oceans is about 1.4x10²¹ liters, with an average concentration of 3.2 parts per billion. This corresponds to 4 billion tonnes of ²³⁸U. On the other hand, the energy content is very dilute, and vast amounts of water would have to be processed to extract uranium.
- Not really feasible!.





Cross-sections – ²³³U



T _{1/2} => 159.25 ky +/- 200.00 y [5.026E+12s +/- 6.312E+09s]

* DECAY MODES:

Alpha decay, possibly followed by Gamma emission Q_value => 4.9089 MeV +/- 0.0012 MeV Branching => 1 0

* NUCLIDE MAT. NUMBER: 9234 The following number of lines are present: Gamma: 116 Alpha: 37 Electron: 235 X-rays: 7





Cross-sections – ²³⁵U



T _{1/2} => 703.81 My +/- 500.01 ky SPIN => 7/2 -

* DECAY MODES:

Alpha decay, possibly followed by Gamma emission Q_value => 4.679 MeV +/- 0.0025 MeV Branching => 1 0 Fission decay, followed by Gamma emission Q_value => 176.4 MeV +/- 4.6 MeV Branching => 2e-10 +/- 1e-10

* NUCLIDE MAT. NUMBER: 9240 The following number of lines are present: Gamma: 49 Alpha: 17 Electron: 127 X-rays: 7





Cross-sections – ²³⁸U



T _{1/2} => 4.47 Gy +/- 5.00 My

* DECAY MODES:

Alpha decay, possibly followed by Gamma emission Q_value => 4.2703 MeV +/- 0.0039 MeVBranching => 0.999999Fission decay, followed by Gamma emission Q_value => 173.6 MeV +/- 3.4 MeVBranching => 5.4e-07 +/- 2e-08

* NUCLIDE MAT. NUMBER: 9249 The following number of lines are present: Gamma: 2 Alpha: 3 Electron: 6 X-rays: 2







Transuranic - TRU



Minor Actinides MA





- Chemical elements of atomic number **greater than 92**. At least 20 such elements identified, more than 100 radioactive isotopes.
- Produced artificially by interactions of heavy atoms either with neutrons produced in nuclear reactors or in specially designed nuclear explosives, or with charged particles from cyclotrons or linear accelerators.
- The first **11 transuranium elements**, together with **actinium**, **thorium**, **protactinium**, and **uranium**, constitute the actinide elements **chemically analogous** to the rare earth elements: **neptunium**, **plutonium**, **americium**, **curium**, **berkelium**, **californium**, **einsteinium**, fermium, mendelevium, nobelium, and lawrencium.



Production and Uses of Transuranium.

The radioactive decay rates of the transuranium elements increase with increasing atomic number;

The very heavy transuranium nuclei, as californium, *fission spontaneously*.

It is extremely difficult to manufacture large quantities of the elements heavier than plutonium

• solution: bombarding uranium and plutonium with very intense fluxes of neutrons in reactors. Quantities produced: several milligrams per year of berkelium, californium, and einsteinium, and small amounts of fermium.

Isotopes such as ²³⁸Pu and ²⁴⁴Cm are used as extremely compact, long-lived sources of power, with the radioactive-decay heat converted directly to electricity by thermoelectric devices.

Transuranium isotopes such as ²⁴¹Am and ²⁵²Cf have medical and industrial uses.





 ²³⁵U – the only mined fissile isotope, most of the reactor concept require enriched uranium!







- The nuclear fuel cycle (NFC) describes the different steps of the nuclear fuels' path from the mining of uranium, through conversion, enrichment and fuel fabrication – which make up the front-end of the fuel cycle, to irradiation of the fuel in the reactors, reprocessing, cooling, storage and finally the disposal – which make up the back-end.
- The fuel cycle is thus all the steps required to supply a nuclear reactor with fuel and take care/make use of the spent fuel. The most common nuclear fuel today is Uranium Oxide (UOX) and Mixed Oxide Fuel (MOX). Other nuclear fuels considered for industrial use are metallic fuel and nitride fuel.











Few slides about the front end of the Nuclear Fuel Cycle











The first steps of the nuclear fuel cycle:

- Mining (depending on the depth at which the ore body is found)
 - surface (open cut),
 - in situ (solution mining)
 - underground mining techniques,
- Milling
 - A uranium oxide concentrate is produced $(U_3 0_8)$, known as yellowcake.
 - The remainders of the uranium ore, tailings, produced in very large volumes. The tailings contain low concentrations of naturally occurring radioactive materials, such as ²³²Th and ²²⁶Ra, which decays to the radioactive gas ²²²Rn.The total radioactivity of the tailings is less than in the original ore.





- At the conversion facility:
 - the yellowcake is first refined to uranium dioxide (UO_2) - can be used as fuel in the type of reactors not requiring enriched uranium (Canadian CANDU and British MAGNOX reactors use natural uranium as fuel).
 - To perform the enrichment the uranium converted into uranium hexafluoride (UF₆) - a solid at room temperature and a gas at slightly higher temperature.
 - The conversion is carried out at conversion plants in Europe, Russia or North America.





Enrichment

- Done by separating the uranium isotopes ²³⁵U and ²³⁸U
 - Two enrichment processes are in commercial use,
 - gaseous diffusion
 - gas centrifuge,
 - Both use the mass difference between ²³⁵U and ²³⁸U to separate the isotopes.
 - Enrichment accounts for almost half of the cost of nuclear fuel and about 5% of the total cost of the electricity generated.
 - Commercial enrichment plants in operation in France, Germany, Netherlands, UK, USA and Russia.
 - Smaller plants in operation elsewhere.
 - The first enrichment plants were built in the USA and used the gaseous diffusion process, but more modern plants in Europe and Russia use the centrifuge process. The centrifuge process has the advantage of using much less power per unit of enrichment and can be built in smaller, more economic units. As the enrichment technology could be used by clandestine weapon programs to enrich weapon-grade uranium for non-peaceful use, the enrichment facilities are one of the most crucial steps of the fuel cycle and subject to proliferation concerns.





- Natural uranium has an abundance (by number of atoms) of
 - 0.0055% 234U,
 - 0.720% 235U,
 - 99.275% 238U.
- Fuel used in LWRs is enriched to ²³⁵U from under 1% to over 4%, with typical enrichments in the neighborhood of 3%.
- This material is known as slightly enriched uranium, in contrast to the highly enriched uranium (HEU) used for nuclear weapons and submarine reactors. HEU starts at 20%
- Within the core of a given reactor, enrichments vary with the location of the fuel assemblies.





- the enriched material itself
- the depleted uranium (sometimes called enrichment tails) Typically, - 0.2% to 0.35% ²³⁵U remaining.
- The depleted U is even less radioactive than natural uranium and is sometimes used in special applications, such as armor-piercing shells, where the high density of uranium is useful ($\rho = 19$ g/cm).





- Gaseous diffusion
- Centrifuge separation
- Aerodynamic processes
- Electromagnetic separation
- Laser enrichment





- The average kinetic energy of the molecules in a gas is independent of the molecular weight M of the gas and depends only upon the temperature.
- At the same temperature, the average velocities are therefore inversely proportional to \sqrt{M} . For uranium in the form of UF₆, the ratio of the velocities of the two isotopic species is 1.0043.
- If a gas sample streams past a barrier with small apertures, a few more ²³⁵U molecules than ²³⁸U molecules pass through the barrier, slightly enriching the gas in the lighter isotope. The ratio of ²³⁵U/²³⁸U before and after passing the barrier is the enrichment ratio u. Its ideal or maximum value is given by the velocity ratio: a = 1.0043.
- Starting with natural uranium (0.71%) and with tails depleted to 0.3% - about 1200 enrichment stages are required to achieve an enrichment of 4%.





Any fluids – liquid or gaseous – are separated by mass in a highspeed centrifuge. It is possible, therefore, to enrich UF_6 using several stages of centrifuges. The power requirement per unit output is much less for centrifuge separation than for diffusion separation.







- Exploit the effects of centrifugal forces, but without a rotating centrifuge. Gas typically, UF₆ mixed with hydrogen – expands through an aperture, and the flow of the resulting gas stream is diverted by a barrier, causing it to move in a curved path. The more massive molecules on average have a higher radius of curvature than do the lighter molecules, and a component enriched in U is preferentially selected by a physical partition. The process is repeated to obtain successively greater enrichments.
- The gas nozzle process was developed in Germany as the Becker or jet nozzle process. A variant with a different geometry for the motion of the gas stream, the so-called Helikon process, has been developed and used in South Africa.





• When ions in the same charge state are accelerated through the same potential difference, the energy is the same, and the radius of curvature in a magnetic field is proportional to. Thus, it is possible to separate the different species magnetically. This separation can be done with ions of uranium, and so conversion to UF_6 is not in principle necessary. Overall, this approach gives a low yield at a high cost in energy, but it has the advantage of requiring a relatively straight forward technology (calutrons).





• The atomic energy levels of different isotopes differ slightly. This effect can be exploited to separate ²³⁵U from ²³⁸U, starting with uranium in either atomic or molecular form. For example, in the atomic vapor (atomic route) laser isotope separation (AVLIS) method, the uranium is in the form of a **hot vapor**. The ionization energy to remove an electron is 6.2 eV. Lasers precisely tuned to the appropriate wavelength are used to excite ²³⁵U atoms, but not ²³⁸U atoms, to energy levels that lie several eV above the ground state. An additional laser is used to ionize the excited ²³⁵U atoms. The ionized ²³⁵U atoms can be separated from the unionized ²³⁸U atoms by electric and magnetic fields. It is not possible to obtain complete enrichment in a single stage. The ionization of ²³⁵U is not complete, and some ²³⁸U is ionized by collisions and is collected with the ²³⁸U.





Most nuclear fuel used in light water reactors is in the form of uranium oxide (UO₂). This is not a single compound but rather a mixture of oxides (UO_n), where n on average ranges from 1.9 to 2.1. The enriched UF₆ is chemically converted into UO₂. The UO₂ is processed into a fine powder and is then compacted and sintered to form rugged pellets. During sintering, the oxygen content of the fuel can be adjusted. The pellets are corrected in size, to close tolerances, by grinding. The pellets are loaded into zircaloy fuel pins that are arranged in a matrix to form the fuel assembly.





Reactor fuel






- The uranium oxide UOX- is the fuel currently in use in LWR. A part may be replaced by Pu containing Mixed Oxide Fuel - MOX.
- Ceramic fuel, a solid and brittle inorganic material.
- UOX chemically stable, has a high melting point - 2865°C.
- The fission products are retained within the UO_2 crystal even at a high burnup.
- UOX has a low thermal neutron capture cross section
- Drawback of a ceramic fuel its relatively poor thermal conductivity





$$\int_{0}^{1} n + \frac{238}{92} U \rightarrow \frac{239}{92} U + \gamma$$

$$\int_{0}^{239} Np + \beta^{-} + v$$

$$\int_{0}^{239} T_{\frac{1}{2}} = 23 \text{ min}$$

$$\int_{0}^{239} T_{\frac{1}{2}} = 2.3 \text{ days}$$

$$\int_{0}^{239} Pu + \beta^{-} + v$$

$$Reactor fuel$$

$$\int_{0}^{239} Pu$$

$$Fissile, T_{\frac{1}{2}} = 24.5 \times 10^{3} \text{ years}$$





0

 $^{233}_{92}U$ **Fissile,** $T_{\frac{1}{2}} = 1.6 \times 10^5$ years





²³⁸U, ²³²Th – X.sections















- Mixture of recovered Pu oxide (PuO₂), with uranium (natural or depleted) oxide and recycled as new fuel.
- can replace some of the UOX fuel elements in the core.
- MOX in LWRs the proportion of plutonium is usually 3-10%.
- MOX fuel, consisting of about 7% plutonium, is equivalent to UOX fuel enriched to about 4.5% ²³⁵U, assuming that the plutonium has about 60-65% of ²³⁹Pu.
- If weapons plutonium is used, consisting of more than 90% ²³⁹Pu, only about 5% plutonium is needed in the mix, due to the higher fission probability of ²³⁹Pu compared to the other plutonium isotopes ²⁴⁰Pu, ²⁴¹Pu and ²⁴²Pu.





- The fuel cladding Zircaloy -surrounds the fuel and serves many purposes:
 - provides the heat in the fuel to be transferred to the coolant,
 - prevents corrosion of the fuel by the coolant.
 - retains the fission products.
 - Does not affect too seriously the neutron economics of the core, - a small thermal neutron absorption cross section. T
 - alloys containing a few percent of the elements tin, iron, chromium and nickel.





- Burnable poisons neutron absorbing materials
 - a necessary ingredient in the fuel to accomplish a more efficient reactivity control and a better fuel utilization.
 - compensate for fuel depletion, fission products with a high neutron absorption cross section and temperature effects that introduce negative reactivity.
 - Chemical compounds of boron (B_4C) and gadolinium (Gd_2O_3) are being used as burnable poison materials.





- a measure of the power outage of the fuel (MWd/kgHM or GWd/tHM).
- defined as the thermal effect, produced during a certain operating time and per unit mass of uranium
- a typical LWR's maximum burnup value is today around 40 GWd/tHM.
- for economical reasons there is an urge to increase the burnup and thus the power outage, as long as it poses no safety risk.





What is happening during burnup:







²³⁹Pu X-sections







End of the "FRONT END"



Trieste-2005

Spent Nuclear Fuel

Initial ²³⁵ U enrichment Fraction of nuclei fissioned Transuranium fraction	3.7% 4.3% 1.2%
²³⁷ Np	4.6%
²³⁸ Pu	2.3%
²³⁹ Pu	49.0%
²⁴⁰ Pu	21.5%
²⁴¹ Pu	11.4%
²⁴² Pu	6.2%
²⁴¹ Am	2.8%
²⁴³ Am	1.6%
²⁴⁴ Cm	0.5%

Spent nuclear fuel consist of three groups of elements:

Uranium still constitute for the main part of the spent fuel. Especially most of the ²³⁸U remains.

Fission products (FP) created through fission reactions make up a few percent of the fuel.

Elements heavier than U – transuranium elements (TRU), formed through neutron capture in the core consist about 1% of the remaining material. Plutonium is the main element in this group.

In average TRU have far longer half-lives than do the FP.





- Recovery of the plutonium + MA from the spent fuel.
- The uranium, plutonium and favourably MA separated from the waste products and fuel assembly cladding, by chopping up the fuel rods and dissolving them in acid.
- The recovered uranium can be returned to the conversion plant for conversion to uranium hexafluoride and subsequent re-enrichment
- The plutonium, as an oxide (PuO₂), converted into MOX fuel.
- Different plans for MA depending on the fuel cycle strategy

• Four plants in the European Union currently produce commercial quantities of MOX fuel; two at the reprocessing facility La Hague in France and one in Dessel, Belgium. The fourth plant is operating at Sellafield in the UK.











Major plants for reprocessing commercial nuclear fuel, in operation or under construction.

Plant	Year of first	Capacit	y (t/yr)
	operation	1994	2005
France			
Marcoule (UPI)	1958	400	0
La Hague (UP2)	1966	800	800
La Hague (UP3)	1990	800	800
India			
Tarapur	1982	~150	~150
Kalpakkam			125
Japan			
Tokai	1981	90	90
Rokkasho-mura	Under	0	800
	construct.		





Major plants for reprocessing commercial nuclear fuel, in operation or under construction.

Plant	Year of first operation	Capacity (t/yr)	
		1994	2005
Russia			
Chelyabinsk	1978	400	400
Krasnoyarsk	Under constr.	0	1500
United Kingdom Sellafield (B205) Sellafield (THORP)	1964 1995	1500 0	1500 ~700





What is the problem??

Radiotoxicity of SPENT FUEL and Sustainability of Nuclear Power



Trieste-2005



Radiotoxicity

Ingestion dose coefficients

Aktinid	ε [nSv/Bq]	КР	ε [nSv/Bq]
²³⁸ U	44	⁹⁰ Sr	0.34
²³⁷ Np	110	⁹³ Zr	0.86
²³⁸ Pu	230	⁹⁹ Tc	0.64
²³⁹ Pu	250	¹²⁶ Sn	5
²⁴⁰ Pu	250	129	110
²⁴¹ Pu	5	131	22
²⁴² Pu	240	¹³⁴ Cs	19
241 Am	200	135Cs	2
²⁴³ Am	200	¹³⁷ Cs	13
²⁴⁴ Cm	120		

By dose coefficients it is meant radioactivity weighted to the biological effect of ingestion of the radioactive material by humans specific for each nuclide.

Radiotoxicity is the sum of the biological effects from all nuclides. It is given in units of e.g. Sv/g.







The radiotoxicity of spent nuclear falls significantly with time.

Dominated by FP radiotoxicity in the short perspective.

¹³⁷Cs and ⁹⁰Sr responsible for the largest part.

TRU becomes the main contributor after some tens of years due to high dose coefficients and long half-lives.

Reference level debateable.





Fissioning heavy nuclides



²³³U, ²³⁵U and ²³⁹Pu are fissile in thermal spectrum, the rest are not.

The fission over absorbtion probability depends on the energy of the neutron causing the reaction.

At high neutron energies (MeVrange), the TRU-nuclides are fissionable.





Transmutation

- the transformation of one isotope into another isotope or element by changing its chemical properties
- first demonstrated by Rutherford in 1919. He transmuted ¹⁴N into ¹⁷O by α -particles from naturally radioactive isotopes.
- A nucleus may be transformed by absorption of a particle, such as α -particles, protons or neutrons, possibly followed by fission.
- the Coulomb barrier surrounding heavy nuclei are too great to permit the entry of particles from naturally radioactive isotopes, the transmutation of heavier elements needs higher energy particles to penetrate the nucleus. The development of particle accelerators opened new possibilities for transmutation experiments and in 1939 the first accelerator-driven transmutation was demonstrated by Cockroft and Walton. They let energetic protons collide with a lithium-target in the reaction
- $^{7}\text{Li} + p \rightarrow {}^{4}\alpha + {}^{4}\alpha$
- The most favourable nuclear process used for transmutation of radiotoxic isotopes is neutron absorption.
 - Neutrons are not repelled by nuclei and interaction cross-sections for many transmutation reactions are sufficiently large. Nuclear reactors, existing and also advanced reactor systems, can be used for neutron transmutation as they constitute an extensive source of neutrons.
- Transmutation, in the context of nuclear waste, is a process which reduces the long term radiotoxicity. Of most concern regarding the disposal of the nuclear waste are the actinides and they are, unlike the fission products, fissionable. They may undergo fission, directly or by absorbing neutrons, followed by beta decay, until a fissile nuclide is formed. Most of them will not fission under a certain threshold energy, but above that threshold their fission cross-section is relatively high





P&T – Partitioning and Transmutation: Chemistry (Physical Chemistry) and Physics



Trieste-2005



Objective is a maximum reduction of radiotoxic inventory of spent fuel (repository source term), if a factor of 100 is to be achieved Pu, Am and Cm have to be transmuted







Efficient transmutation of TRU nuclides requires:

- Fast spectrum •
- Uranium free fuel





Consequences of utilising a fast spectrum:

- No moderation water ruled out as coolant candidate, possible options include Pb, LBE, Na and He.
- Material selection to minimise moderation
- Hostile radiation environment pose material problems

(LBE – Lead-bismuth eutectic)







- Low delayed neutron fraction the high delayed neutron fraction in LWRs arise from fission of ²³⁵U
- No or very much reduced Doppler the Doppler effect is very strong in ²³⁸U. Introduction of Am further worsens the situation.

Taken together, these two effects make the reactor – more or less – uncontrollable.

This raises the need for a subcritical core in order to incinerate TRU-based fuels.





- Reactor safety is based on few very important phenomena:
 - Criticality based on delayed neutrons:
 - ensures transient behaviour with managable time constants. Limits: delayed neutron fraction, reactivity must never exceed delayed neutron fraction (β) $\rho << 640$ pcm (0.0064)
 - Reactivity temperature feedbacks MUST be negative
 - Feedbacks privided by physics:
 - Doppler effect
 - Thermal expansion
 - X-section neutron energy dependence





$$P(t) = n_0 \left\{ \frac{\beta}{\beta - \rho} e^{\left(\frac{\lambda\rho}{\beta - \rho}t\right)} - \frac{\rho}{\beta - \rho} e^{\left(-\frac{\beta - \rho}{\tau_p}t\right)} \right\}$$

If $\rho > \beta$

$$\therefore T = \frac{\tau_p}{\rho - \beta} \quad prompt \ critical \ reactor$$

If ρ << β $T = \frac{0.083}{\rho}$





- Origin: broadening of resonances.
- Negative if DOMINATED by CAPTURE resonances, e.g. in ²³⁸U
- Comes into play quickly, reversing (IF NEGATIVE!!) the rise in power output within less than 0.1 second.
- The fuel temperature feedback is not automatically negative in all types of reactors. If a fuel has relatively little ²³⁸U and is primarily made of fissile material, then the main effect of Doppler broadening is to increase the rate of fission at nonthermal energies, giving a positive feedback. Thus, to keep the fuel temperature feedback negative, the fraction of fissile fuel in e.g. liquid-metal fast breeder reactors must be below 30%.













GOAL: maximum net consumption of TRU (and FP)



limited content or no U-238 \rightarrow no *breeding*

fast reactor spectrum \rightarrow high *fission-to-absorption ratios*

Problems to solve: parameters

deterioration of neutronic and kinetic

- moderator void and temperature coefficient
 - fuel temperature coefficient (Doppler constant



- (effective) delayed neutron fraction (β , β_{eff})
- neutron lifetimes (τ)





Pu may be managed/transmuted in critical reactors (mainly fast)

BUT

- Am lacks Doppler feedback, increases void coefficient
- Am fraction in (fast) critical reactors limited to < 5%

So

- Excess Am (& Cm) must be burned in "dedicated" cores
- No Doppler feedback & small βeff requires sub-criticality
 Ergo

Safety and efficiency require subcriticality! Possible radiotoxic inventory reduction factor: 100 – 150





ADS Principal Components







- Power: 800 MW
- Sub-criticality level: $k_{eff} < 0.97$
- Proton accelerator: 30 MW (30 mA, 1 GeV)
- Spallation target: PbBi
- Coolant: Lead-bismuth eutectic or Helium
- Fuel: Pu-, Am-, Cm-oxide or nitride







NUCLEAR FUEL CYCLE BACK END and RECYCLING
















"Triple" Strata













Swedish Reactor Fleet

- 9 BWR and 3 PWR
- 40 years of operation
- 146 TWh total electricity production (2004)
- 75 TWh from nuclear energy
- Once-through fuel cycle
- ~10000 tons of spent fuel produced by 2025









ONCE-THROUGH NFC – Swedish Example















Trieste-2005

ONCE-THROUGH NFC – Swedish Example - Future



Kapsbil



Djupfani.avi



Forvar_tot



Official Swidish Nuclear Fuel Cycle Policy







- ... is a pretty complicated endeavor. A lot of studies have been done, a lot of projects have been devoted to this research:
 - Most recent European Project RED-IMPACT Impact of Transmutation and Reduction/Conditioning of Nuclear Waste on Final Waste Repository. Final report will be delivered in March 2007!
 - A very good Status Report was recently published by the Swedish SKB, "Partitioning and Transmutation. Current developments – 2004", Report TR-04-15, http://www.skb.se





BUT..... First estimations are easy...





- Transmutation/reactor system burns 100kg/year of Heavy Metals in 300 MWth of power
- Feasible burnup of the fuel is 5% of HM for "regular" systems and up to 20% in very advanced systems (impossible in critical reactors), it means that we need an inventory 5 to 20 times larger than an annual consumption, i.e. 500 to 2000 kg for 300 MWth of power.
- Having inventory of 1000 of tons (10⁶ kg) of HM we shall need MANY, MANY, MANY reactor/ADS*years to get rid of this inventory.
- "In equilibrium" we need about 1 ADS per 4-5 LWRs or 1 per ~20 Fast Reactors to burn produced HM wastes
- BUT AT THE SAME TIME WE PRODUCE AND DELIVER ENERGY. NUCLER WASTE DOES NOT NEED TO BE VIEWED ONLY AS A LIABILITY.





Impact of P&T on the Final Repository

Prof. Waclaw Gudowski KTH, Stockholm wacek@neutron.kth.se





Detailed analysis of the fuel cycle





- Schemes based on current industrial technology and possible extensions
- Schemes with partially closed fuel cycles
- Schemes involving fully closed fuel cycles





Schemes based on current industrial technology and possible extensions







Schemes with partially closed fuel cycles





Schemes with fully closed fuel cycles







- Reactor and fuel cycle characteristics
 - PWR-UOX

EPR-type pressurised water reactor with uranium oxide fuel.

- PWR-MOX
 Same reactor with 100% MOX fuel.
 Variant with uranium-plutonium-neptunium mixed-oxide fuel (MOX-Np).
- (PWR-)MOX-UE (Schemes 2a and 2b) As PWR-MOX, but with enriched instead of depleted uranium. Variant with different plutonium content and different fissile-plutonium and uranium enrichment (MOX-UE1), variant with admixed americium (MOX-UE2).
- CANDU (Scheme 1c) CANDU heavy water reactor with DUPIC fuel.

 FR-MOX (Schemes 3cV2, 2c and 2cV) EFR-type sodium-cooled FR with minor-actinide loaded MOX-fuel (Scheme 3cV2).

Plutonium burning variant (*FR-MOX1*) with pure MOX fuel, with or without irradiation positions for americium target assemblies (Schemes 2c and 2cV).





- Reactor and fuel cycle characteristics –cont.:
 - FR-HBU (Scheme 3b) EFR-type plutonium burner with a high burn-up core. The plutonium content of the MOX fuel is increased to 44% to maximise the plutonium consumption.
 - FR-metal (Scheme 3a) ALMR-type sodium-cooled actinide burner with a metal-fuelled, low conversion ratio core. The reactor model is the 600 MWe, metal-fuelled, multiple recycle burner core
 - FR-carbide (Scheme 3cV1) Gas-cooled fast reactor (GCFR) with solid solution carbide fuel as proposed for Gen-IV.
 - ADS-MA (Scheme 3b) Small ADS with a lead-bismuth cooled core with nitride fuel, following a minor actinide burner concept.
 - ADS-TRU (Scheme 3bV) Accelerator-driven, lead-bismuth cooled TRU burner with nitride fuel.





Reactor design and fuel cycle parameters

Reactor	Thermal power (MW)	Net electric power (MW)	Electrical efficiency (%)	Fuel	Fuel burn-up (GWd/tHM)	Storage/ cooling time (a)
PWR-UOX	4250	1450	34.1	UOX	60 ¹	2 / 5 ³
PWR-MOX	4250	1450	34.1	MOX	60 ²	2 / 5
MOX-EU	4250	1450	34.1	MOX-EU	60	2 / 5
CANDU	2159	713	33.0	DUPIC	15	2 / -
FR-MOX	3600	1450	40.3	MOX	140	2 / 5
FR-HBU	3600	1450	40.3	MOX	185	2 / 5
FR-metal	1575	600	38.1	78 Ac- 22 Zr	140	1 / 2
FR-carbide	2400	1158	48.3	(U,Pu)C-SiC	100	2 / 5
ADS-MA	377	119	31.6	²⁹ AcN- ⁷¹ ZrN	150	1 / 2
ADS-TRU	850			AcN-ZrN	150	1 / 2





Heavy metal mass flow and equilibrium fuel composition

Reactor	HM mass flow (kg/TWhe)	Fuel composition after irradiation (%)					
(Scheme)		U	Np	Pu	Am	Cm	
PWR-UOX (1a)	2050	98.5	0.10	1.35	0.08	0.01	
PWR-MOX (1b)	225	91.9	0.02	7.24	0.68	0.16	
MOX-Np (1c)	215	90.4	0.42	8.35	0.71	0.14	
MOX-UE (2a)	575	89.5	0.05	9.32	0.89	0.22	
MOX-UE1 (2b)	238	91.3	0.06	7.74	0.71	0.18	
MOX-UE2 (2b)	711	92.7	0.09	6.05	0.84	0.34	
CANDU (1d)	1997	99.0	0.06	0.87	0.04	0.01	
FR-MOX (3cV2)	890	77.6	0.12	21.12	0.88	0.29	
FR-MOX1 (2c, 2cV)	390	78.6	0.07	20.71	0.55	0.05	
FR-HBU (3b)	106	57.1	0.06	39.81	2.56	0.51	
FR-metal (3a)	289	69.8	0.65	26.60	2.00	0.98	
FR-carbide (3cV1)	849	79.3	0.12	19.48	0.87	0.24	
ADS-MA (3b)	46	5.4	6.09	47.58	23.15	17.72	
ADS-TRU (3bV)	117	1.9	3.29	73.48	12.37	8.96	
Am targets (2c)	0.45	1.4	0.14	43.13	9.78	45.57	







Heavy metal mass flow and equilibrium fuel composition







TRU Inventories







• The multi-recycling of fuels in the Family 2 and 3 schemes leads to high in-pile and outof-pile TRU inventories. These have safety implications due to their high radiotoxicity and influence the time constants under transient conditions, e.g., in the deployment and shutdown phases of a nuclear energy scenario. **Difficult but Managable**





- By fully closing the fuel cycle, the heavy metal content of the waste can be reduced by more than three orders of magnitude, compared with the once-through fuel cycle. Being dominated by the fission products, the volume of the high-level waste will, however, not be noticeably affected by this reduction.
- Due to the higher electrical efficiency of fast reactors, a change from an all-LWR to an all-FR reactor strategy would reduce the fission product mass, and hence the high-level waste volume, by about 30%.
- Only transmutation strategies with fully closed fuel cycles can meet the hundredfold TRU reduction goal of P&T. Partially closed fuel cycles, which cannot achieve such high TRU reductions but are easier to implement, are also useful for managing plutonium and minor actinides. When a transmutation strategy is phased out, it has to be ensured that the TRU waste reduction achieved in the equilibrium phase is not jeopardised by the disposal of the residual TRU inventory.





Conclusions - 2

- The high decay heat and neutron emission of multi-recycled fuels have consequences for the reprocessing and the handling of the fuel. As to the reprocessing, improvements to the flowsheets of aqueous processes and the introduction of pyrochemical methods will be necessary. The pyrochemical methods produce wastes with unusual characteristics which have to be conditioned and disposed using novel waste management methods.
- Since the actinides are mostly recovered, minor actinide management techniques reduce the total decay heat of the waste. While the achievable heat reduction at normal disposal time (50 years) is modest - factor 2 to 4 depending on the scheme- there exists a significant heat reduction potential for longer cooling times. For example, a 20- to 30-fold heat reduction relative to the once-through fuel cycle could be achieved with a cooling time of 200 years.
- Pure plutonium flows being as a simple indicator for the proliferation risk -can be divided into three groups with no, relatively low, and 3 to 4 times higher, mass flows of pure plutonium. Measures for reducing these flows may be coupled with technical or economical drawbacks.
- As to the natural uranium demand, the reduction potential of advanced LWR strategies is about 10%, compared with the once-through fuel cycle. A significant (40%) uranium saving can be achieved with the DUPIC fuel cycle. Higher savings are possible by incorporating fast reactors into the park. An all-FR strategy would allow the uranium demand to be reduced by more than two orders of magnitude.
- All reactor parks including LWRs produce significant amounts of residual uranium. The longterm storage or disposal of this uranium has to be considered as an integral part of the waste management.





Swedish Reactor Fleet

- 9 BWR and 3 PWR
- 40 years of operation
- 146 TWh total electricity production (2004)
- 75 TWh from nuclear energy
- Once-through fuel cycle
- ~ 10000 tons of spent fuel produced by 2025











• Equilibrium Scenario

LWRs and ADS operating in equilibrium in order to assess the ratio of ADS over LWR needed to stabilize TRU production

• Phase-out Scenario

"as soon as possible" reduction of Swedish spent fuel with burning of americium and plutonium in ADS





- Reference Scenario: Swedish LWR park with a once-through fuel cycle
- Equilibrium Scenario #1: LWR + 4 ADS
- Equilibrium Scenario #2: LWR + 4 ADS + MOX LWR
- Phase-Out Scenario #3: LWR + 3 ADS
- Phase-Out Scenario #4: LWR + 4 ADS





- NFCSim developed at Los Alamos
 - written in Java
- Origen 2.2 burn-up/reactivity
 - calculations adjusted with "own" Xsection library (MCB or Monteburns)
- "Transient" effects of a reactor fleet evolving in time
- Reactor types: LWR, FR, ADS





- The Swedish LWR input-data are virtual data of existing reactors
- ADS data are based on KTH subcritical core
- ADS start-up in 2030 (equilibrium and phase-out scenarios)
- MOX-recycling (in equilibrium scenario) in classical LWRs with start in 2000





Swedish EWxample - Result Equilibrium Scenarios



Plutonium inventory per total produced thermal energy

Scenario # 4 ADS118 tons (58%) reductionScenario # MOX + 4ADS195 tons (69%) reduction







Americium inventory



Americium inventory per total produced thermal energy

Scenario # 4 ADS23 tons (79%) reductionScenario # MOX + 4ADS19 tons (66%) reduction













Americium and Neptunium Scenario #3 (LWR + 3 ADS) Americium and Neptunium Scenario #4 (LWR + 4 ADS)

Hur kan mer Am ha föbrukats i scen 4ADS??

Scenario # 3 ADS Scenario # 4 ADS 4.4 tons (63%) reduction 4.6 tons (66%) reduction





• Long-term Equilibrium Scenario:

4 ADS units of 800 MWt are sufficient to burn the amount of plutonium and americium corresponding to their buildup during a continuous operation

• Phase-Out Scenario:

3 ADS is optimal for an "as soon as possible" reduction of Sweden's accumulated amount of spent fuel




- Advanced technological approach needed
 - Reprocessing
 - Fuel fabrication
 - ADS
 - HLW management
- Multinational/Nordic waste management cooperation might be an option
- Public acceptance





Sustainable Development of Nuclear Energy: the Role of Accelerator Driven Systems In collaboration with M. Salvatores CEA/ANL

Outline:

- 1. Fuel cycle strategies and Accelerator Driven Systems
- 2. ADS-based transmutation: dedicated fuels and fuel processing
- 3. ADS-based transmutation: deployment strategies
- 4. ADS-based transmutation: a « regional » approach
- 5. A path towards a transmuter ADS
- 6. Conclusions and perspectives





- Pu is a good nuclear fuel energy production
- Pu is a source of surplus neutrons breeding
- Pu is a radiotoxic materials in nuclear waste
- ²³⁹Pu is a nuclear weapon material a proliferation risk





Double strata fuel cycle, within a continuous use of nuclear energy, and Pu being a resource:

- → MA (and some Pu) are "segregated" in a specific fuel cycle, with no impact on the commercial fuel cycle (where most of the Pu is recycled). This option can be an alternative if the introduction of Gen-IV FR with multiple recycle of notseparated TRU (which is the optimal strategy in order both to minimize wastes and to provide a sustainable development of nuclear energy), is delayed.
- \rightarrow The optimal Pu/MA ratio is close to one, according to several studies.







Open fuel cycle (or even in a nuclear phase-out perspective)

- In order to reduce the burden on a final geological storage.
- All TRU have to be considered. Pu is the dominant component: Pu/MA~4/1.

Storage of nuclear wastes







Double strata »: Pu still a resource. Gen-IV FR deployment delayed Details







Reduction of Pu+MA stockpile (Pu considered as waste)



Fuel fabrication

• Reprocessing

If timeframe for reducing stockpile ~100 y, ~20% of initial stockpile is not burnt.





- In both cases ("double strata" and "open cycle"), <u>subcritical</u> dedicated "transmuters" (ADS) offer advantages in terms of safety and flexibility to manage a wide range of different fuels.
- However, recent studies in the US have indicated the potential feasibility of deep burner <u>critical</u> fast reactors in the "double strata", with conversion ratio CR<0.25, i.e. with U/Pu/MA ~ 0.35/0.55/0.1.
- To examine the viability of the dedicated ADS deployment, it is necessary to establish:
 - A. Dedicated fuels and fuel processing.
 - **B.** Needs in terms of number and pace of ADS deployment.

These two points will be analysed in what follows.





ADS-based transmutation: dedicated fuels and fuel processing

Needs and characteristics of dedicated (U-free) fuels, spent dedicated fuel reprocessing and dedicated fuel fabrication facilities.

Fuels	Fuel fabrication and Fuel processin performance		Impact on repository
U-free fuel: Pu/MA~1/14/1 -To be developed and validated -Very high He production -Transient behaviour to be assessed Experiments performed on different fuel types: metal: USA; oxide: Europe; nitride: Japan,USA,Europe Different matrix explored: for oxide, candidates are: MgO or ZrO ₂ (CERCER); Mo (CERMET) for nitride	 Fabrication: High neutron doses: Remote Need new processes: Vibrocompaction? Pyrometallurgy? Performance under irradiation: Very limited experience with (Pu, Am)-bearing fuels No experiment with Cm 	To be developed Inert matrix choice needs pre-screening: Can have impact on - reprocessing flow-sheet - secondary wastes Pyrochemistry? -Cm behavior in molten salts?	 -Losses at reproces. (Pu): 0.1% -Losses at reproces. (Pu+MA): 0.1%? -Secondary waste (according to matrix) If total TRU losses ~0.1%, masses, heat load, radiotox. reduced by ~100 -MgO matrix can have impact on final waste form (glass)
(TRU,Zr)N best candidate (?)			





- For <u>ADS deployment scenarios</u>, two examples can help to quantify needs:
 - The case of a Country A which considers the phase-out of nuclear energy, but looks for a minimization of wastes to be sent to a final repository. In the case considered, the amount of Pu and MA built-up at an hypothetical phase-out date ~2025 are respectively : $M_{Pu} \simeq 120$ t and $M_{MA} \simeq 20$ t.
 - The case of a Country B with a LWR fleet producing ~ 60 Gwe.
 MOX fuel is multirecycled in the LWR with a specifically optimized core fuel assembly. MA are separated and sent, together with some Pu, to the dedicated transmuters of the second stratum in a "double strata" strategy





The ADS is a Na-cooled, metal fuelled facility with an LBE target. Eight 840 MWt facilities deployed in the first generation and three in the second.

Top-Level ADS Parameters

Target K _{eff}	0.97 (BOC)	
	0.94 (EOC)	
Core Inventory	3000 kgIHM	
Thermal Power	840 MWt	
Discharge Burnup	200 MWd/kg	
Fuel Mangt	5 batches / core	
Cycle Time	168 days	

Facilities Deployed







Accelerator driven systems are deployed beginning in 2030.

Same top-level ADS design parameters. The more favorable feed stream, however, allows a 2 batch/core reloading scheme with the charge and discharge k_{eff} values being held at 0.97 and 0.94 respectively.







- The number of ADS to be deployed is relatively significant (e.g. of the order of 7-10% in terms of total power, in the double strata case), and with a relatively aggressive pace of deployment.
- Moreover, in the case of Country A which wants to burnTRU, after ~ 100 years the TRU inventory not yet burnt-out, is still significant (~ 20% of the initial inventory), despite the large number of facilities (ADS and fuel cycle) deployed.
- These features have suggested a "regional" strategy, e.g. the two countries can cooperate, both to reduce the number of fuel cycle facilities and the ADS to be deployed, with mutual benefits.

A NEW PARADIGM!!!





Regional Blend and Burn Scenario: Flowchart







A symbiotic relationship between Country B MA and Country A SNF.

ADSs are deployed beginning in 2030; their rate of deployment is governed by the inventories of SNF and MA available to start up the first facilities.

In 2100, 20 facilities of 840 MWt (~275 MWe) capacity are operational.







	Country A	Country B	Regional Scenario (A+B)	Difference:
Number of 840 MWt ADS deployed	8 (2040-2070) +3 (2070-2080)	16 (after 2085)	20 (after 2095)	7

Moreover in the Regional Scenario:

- Only one Reprocessing/Fuel fabrication Facility and, potentially, only one Repository for the Two Countries

- Country A has no leftover inventory from ADS (as it is the case if ADS deployed only in that country): all SNF reprocessed by 2050.





- An ADS-based transmutation strategy does require the demonstration of the feasibility of:
 - Dedicated fuel cycle facilities: dedicated (i.e. with high MA loading) fuel fabrication and fuel reprocessing facilities.
 - ADS concept (safety, operation, ad-hoc instrumentation, coupling of components: high power proton accelerator with stringent requirements on reliability; spallation target; subcritical core).
- As for the ADS concept demonstration at power, there is the possibility of an interesting synergy with the perceived need to deploy in Europe (beyond ~2010 i.e. after the PHENIX shut-down) a new fast spectrum irradiation facility, dedicated to the Gen-IV innovative fuel development.
- After a first phase, devoted to basic coupling experiments (~2005 2010) a second phase can be envisaged, focussed on a "dual" purpose facility.





- The issues of ADS-based transmutation, after ~15 years of research, are now better understood
- Most of the research has been made in support of the ADS concept itself, and of the two key associated technologies: HPPA and spallation targets, which benefited from intense neutron source research.
- Much less is known in the crucial field of dedicated U-free fuels which have to be developed in a real life ADS-based transmutation strategy. Even less is known in the equally crucial field of reprocessing methods for irradiated dedicated fuels.
- This situation suggests the need for future (probably expanded) collaborative programs, to set up a more balanced share of activities and to focus on issues which have not been dealt with up to now and which can present significant difficulties (e.g. Cm management).
- As far as the ADS demonstration at power, one can argue that the cost of such facility and the relatively limited time needed for demonstration purposes, could suggest the deployment of a dual purpose facility, i.e. a robust fast spectrum irradiation facility with a mission limited in time of ADS demonstration at power.





- As far as the ADS-based transmutation, it seems difficult to foresee its implementation by a country in isolation, in particular if the ADS-based transmutation is proposed in the frame of spent fuel legacy minimisation.
- Synergies among countries (with different policies), in a regional context, can offer a more favourable and credible perspective, as shown in the example presented in this paper. A further extension of the proposed regional approach (e.g. at the European level) could help to clarify issues and to show a path forward.





Reference scenario for a sustainable development of nuclear energy with waste minimisation







"Triple" Strata







Are there any other options??



Trieste-2005