Thorium for Nuclear Energy – a Proliferation Risk?

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Outline

- Thorium / Uranium Cycle
- Uran-233 as weapon usable material
- Proliferation resistance of U-233 / Uranium-232 contamination
  - Possible solutions and their limitations
- Conclusions

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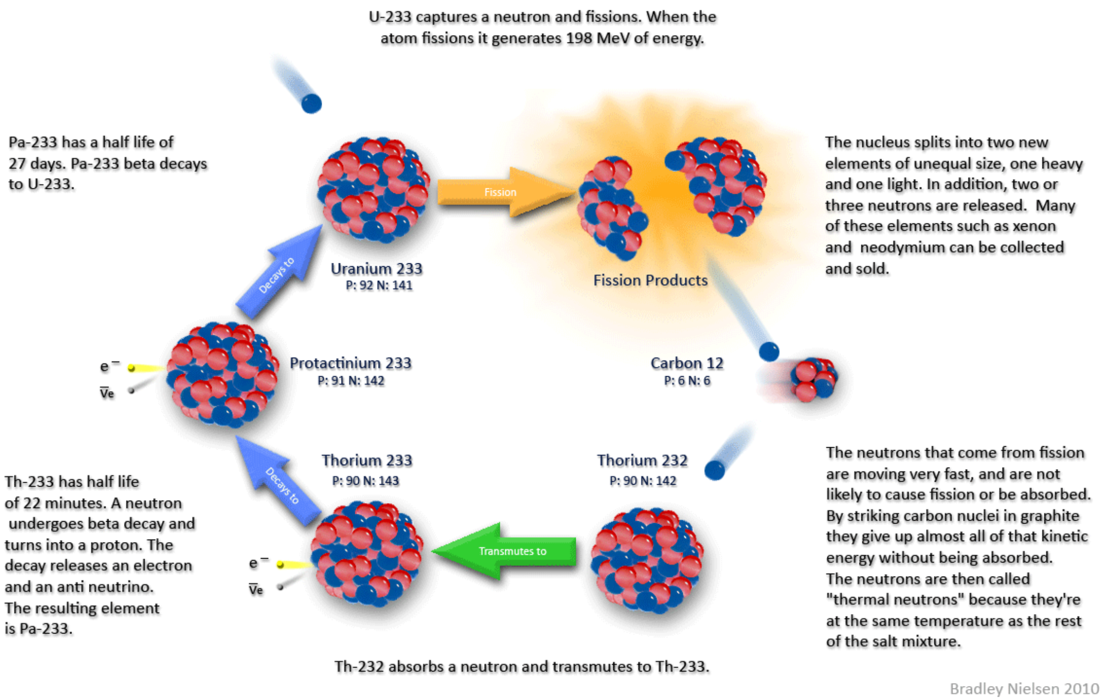
A brief depiction of the thorium cycle



- Thorium-232 as fertile and uranium-233 as fissile material
- More then two neutrons per fission of uranium-233 in wide energy range
- Breeding ratios even with thermal reactors, in opposition to U238-Pu239 cycle

Quelle Bild: http://energyfromthorium.com
Urheber: Bradley Nielsen 2010
Quelle Text: http://www.homelandsecuritynewswire.com/dr20120916-thorium-to-play-limited-role-in-u-k-future-power-supply

Thorium dioxide pellets for reactor operation

Thorium dioxide pellets could be the nuclear fuel of the future if proliferation concerns can be addressed.

Source picture: http://www.nature.com/nature/journal/v492/n7427/full/492031a.html

SOURCE: URANIUM 2011: RESOURCES, PRODUCTION AND DEMAND (OECD NEA/IAEA; 2012)

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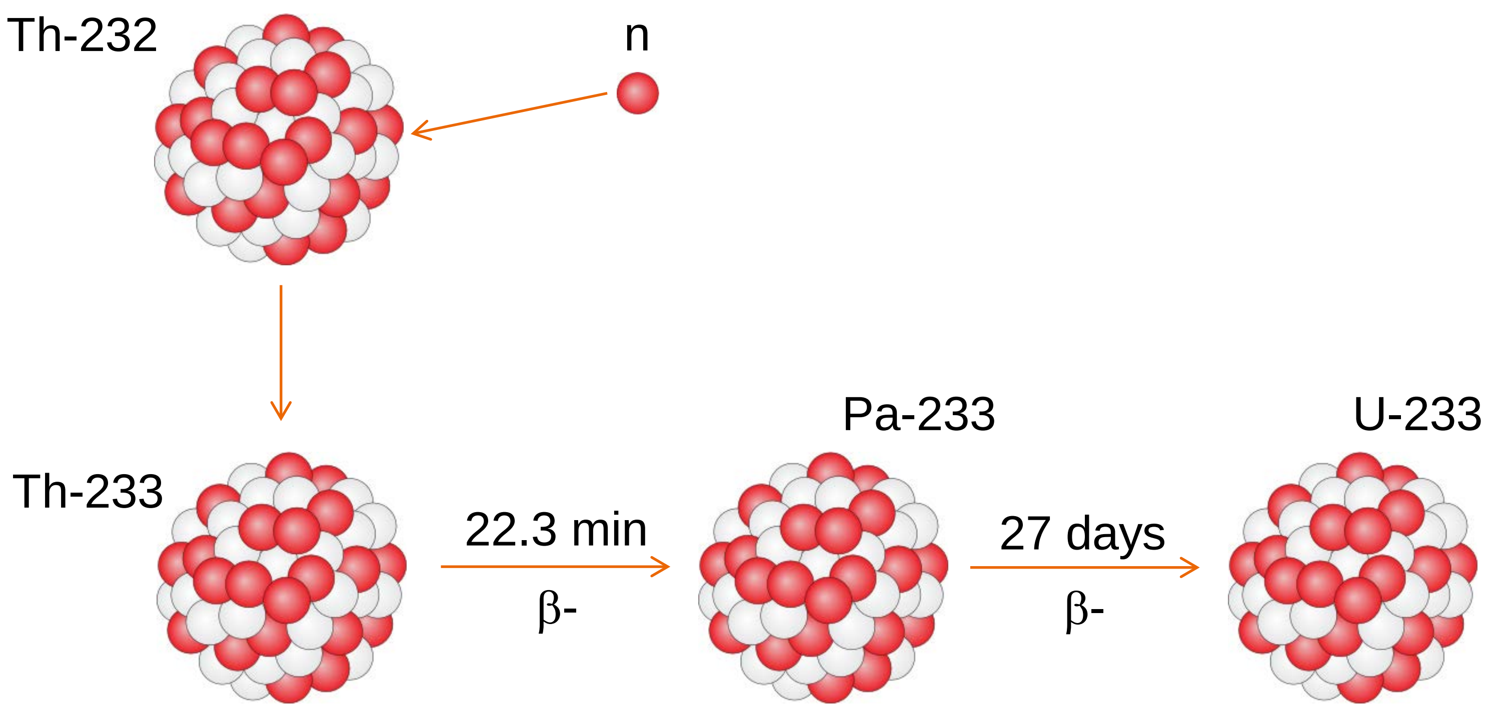
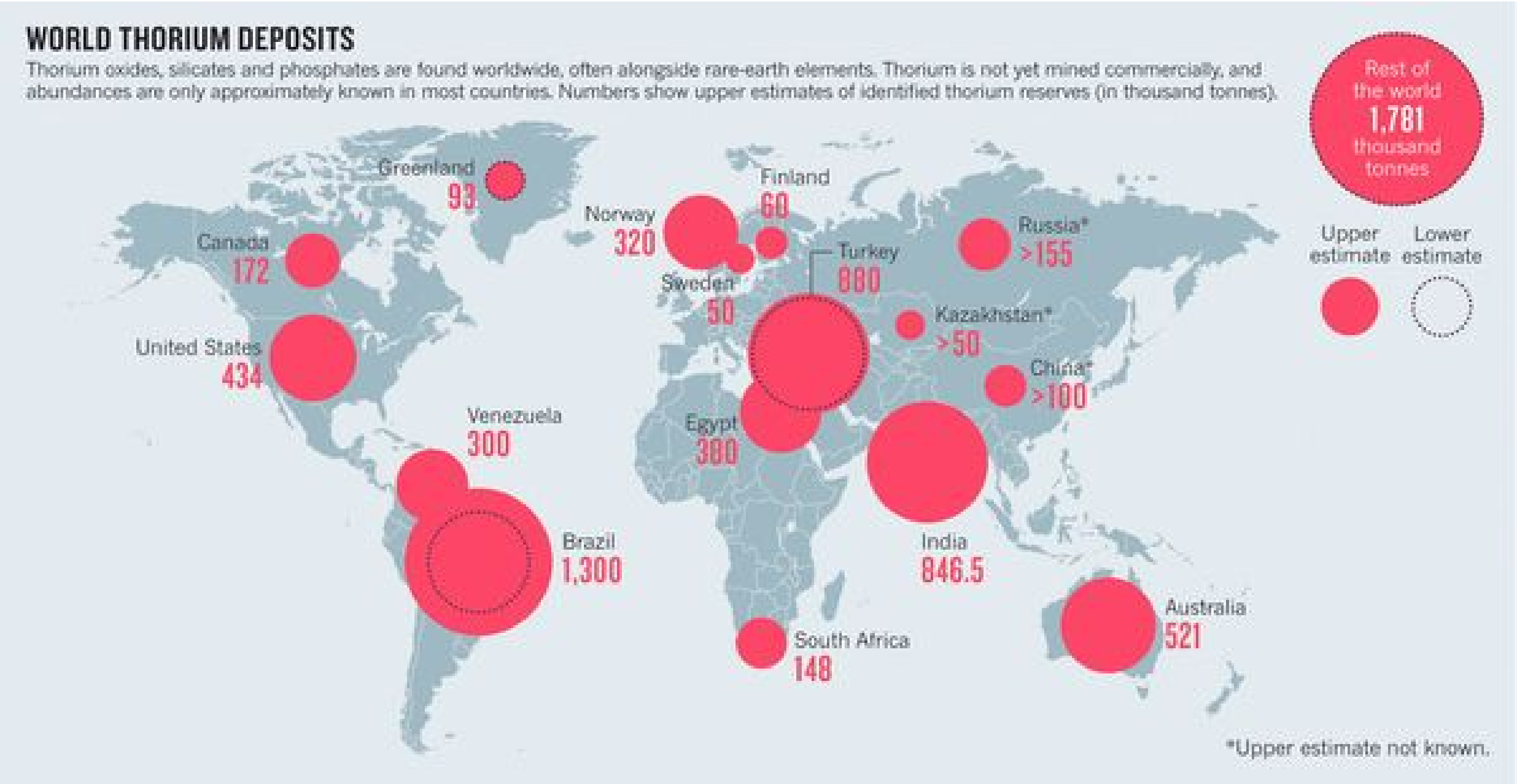
Breeding reaction of Thorium / Uranium Cycle



World Thorium Deposits

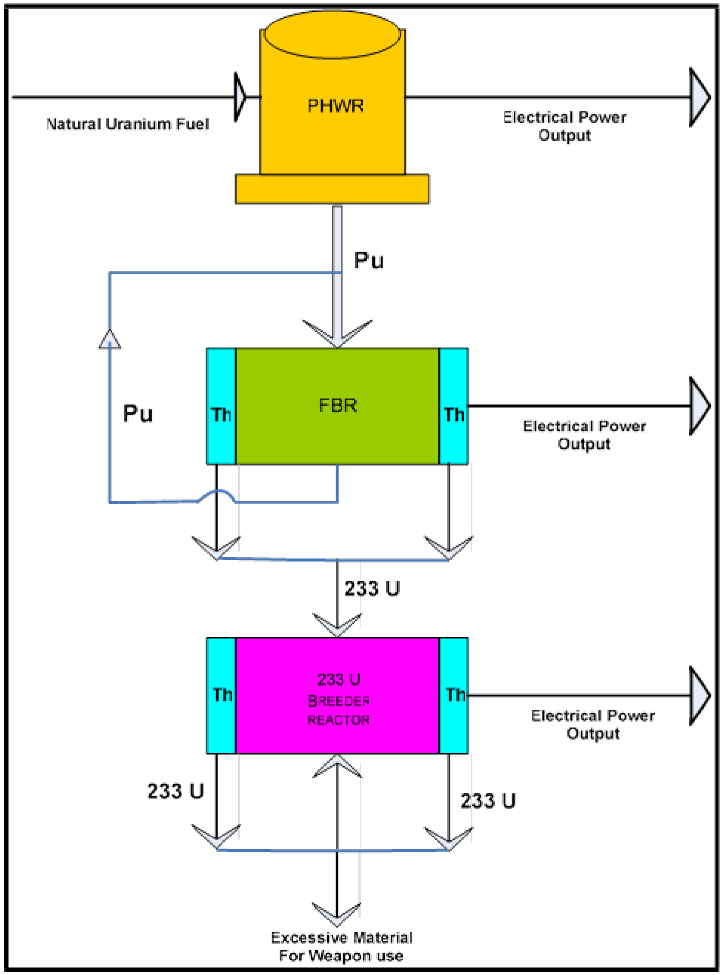
Thorium deposits more than five times higher as of Uranium



Source picture: http://www.nature.com/nature/journal/v492/n7427/full/492031a.html

SOURCE: URANIUM 2011: RESOURCES, PRODUCTION AND DEMAND (OECD NEA/IAEA; 2012)

India's three-stage nuclear power program



- Stage 1
  - Natural Uranium in heavy water pressurized reactor → Pu 239
- Stage 2
  - Fast breeder with MOX Fuel. If enough Pu-239 is generated, Thorium is used in blanket
- Stage 3
  - Thermal breeder (AHWR) with Thorium-Uranium cycle
- Stage 1 and 3 are startup stages for 3

source: A Technical Study of Nuclear Fuel Cycle and the Three Stage Power Program of India for US-India Nuclear Cooperation, p. 1 - 2

Properties of different fissile material

		U-233	U-235	Pu-239
Half life	years	1.6×10^5	7.0×10^8	2.4×10^4
Fertile material		Th-232 (9.6 ppm)	Nat U	U-238 (2.7 ppm)
Breed cross-section	Barn	7.4		2.7
Fission neutrons per absorption	1MeV	2.5	2.3	2.9
	25 meV	2.28	2.07	2.11
Critical mass	kg	15	52	10
Spont. fission	$(s*kg)^{-1}$	0.5	0.6	2.5×10^4

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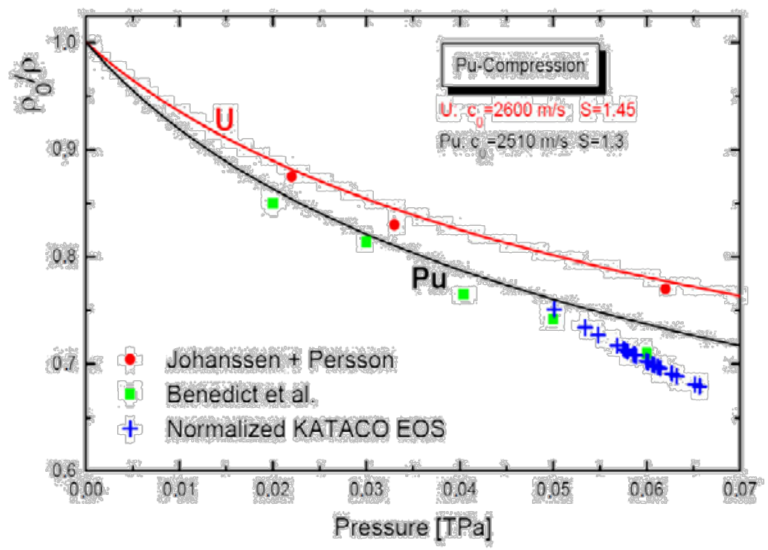
Desirable Features of Thorium cycle

- Th-232 / U-233 generate less minor actinides than U-238 / Pu-239, waste is less radiotoxic, Waste burner
  - Valid only in open or few cycles
    with repeated reprocessing MA could build-up
- Thorium can be processed like Uranium and Plutonium.
- ThO_2 is highest possible oxidation state. No further oxidation could occur like e.g. $UO_2 \rightarrow U_3O_8 \rightarrow UO_3$
- ThO_2 extremely stable
  - Higher melting point (3300°C) than UO_2 (2900°C) and PuO_2 (2400°C)
  - Higher thermal conductivity
  - Higher retention of (volatile) fission products
  - **But**: stability cumbersome for reprocessing e.g. boiling TOREX solution

Uran-233 as nuclear weapon material

		U-233	U-235	Pu-239
Critical mass	kg	15	52	10
Spont. Fission	$(s*kg)^{-1}$	0.5	0.6	$2.5x10^4$
Fission xsection	Barn	1.95	1.24	1.80
Contaminant		U-232	U-238	Pu-238 Pu-240 Pu-241 Pu-242

- At least two nuclear test known, that employed Uranium-233 as fissile material
- USA: 1957, Teapot MET, 22 kt
- India: 1998, Shakti V, 0.2 kt



Bildquelle G.Kessler Proliferation Proof Uranium and Plutonium Fuel Cycle KIT Scientific Publishing 2011

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Activity and neutron emission

Nuclide	Half life	Specific activity	Spontanous fission neutrons
U-232	69 a	$8.20 * 10^{11}$ Bq/g	**$1.7 * 10^{+3}$ n/(g*s)**
U-233	$1.62 * 10^{5}$ a	$3.10 * 10^{8}$ Bq/g	$1.1 * 10^{-3}$ n/(g*s)
U-235	$7.10 * 10^{8}$ a	$7.77 * 10^{4}$ Bq/g	$6.0 * 10^{-4}$ n/(g*s)
U-238	$4.51 * 10^{9}$ a	$1.23 * 10^{4}$ Bq/g	$1.1 * 10^{-2}$ n/(g*s)
Pu-239	$2.41 * 10^{4}$ a	$2.27 * 10^{9}$ Bq/g	$2.2 * 10^{-2}$ n/(g*s)
Pu-240	$6.58 * 10^{3}$ a	$8.36 * 10^{9}$ Bq/g	**$1.0 * 10^{+3}$ n/(g*s)**
Np-237	$2.14 * 10^{6}$ a	$2.60 * 10^{7}$ Bq/g	$1.0 * 10^{-2}$ n/(g*s)

- Additional neutron background from (α,n) reaction on light elements from highly active Pu-240 and U-232
- U-232 only in **ppm**, Pu-240 in **percent** content in WG-material
  → Influence in U-233 1 to 3 orders of magnitude less than Pu-239

Pros and Cons of weapon grade material (for nuclear explosives)

U-233	U-235	Pu-239
▪Strong gamma emission ▪Breeding / reprocessing	•Large critical mass •Enrichment technology	•Neutron Background •Breeding / Reprocessing •Radiotoxic
•Small critical mass •High isotopic purity •Higher burnup possible •Gun Type???	•Low neutron background •No breeding / reprocessing •Gun-Type possible	•Very small critical mass •High compressibility •$\delta \rightarrow \alpha$ phase transition

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Significant Gamma-Energies (E) und Intensities (I) of Nuclear Fission Material

Material	E [keV]	I [γ/g. sec]
U-233	291.3 317.2	56 000 81 000
U-235	185.7	42 000
U-238	1001.0	100
Pu-239	375.0 413.7	36 000 34 000
Pu-240	160.3 212.5	33 000 2 400
Np-237	95.9	670 800
Am-241	59.4	46 670 000
Am-242^{m}	14.6	107 640 000
Am-243	749.7	4 884 000 000

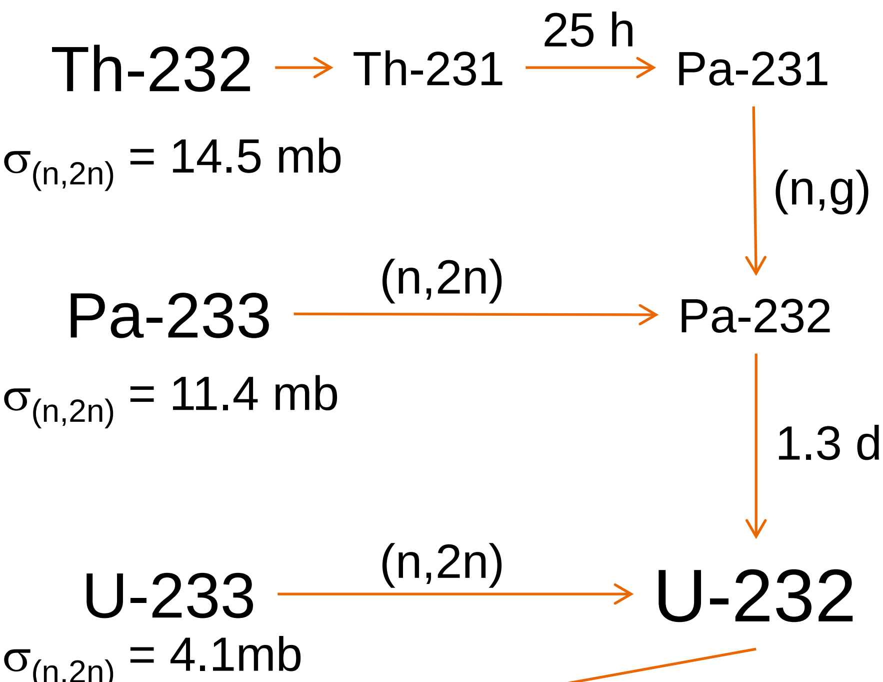
Factors concerning the weapon usability of U-233

- Breaded U-233 contains U-232 only on ppm scale, is intrinsically pure fissile material without the need to develop enrichment technology.
- Critical mass and cross sections more similar to Pu-239 than U-235
- Neutron background less than Pu-239, less sophisticated implosion design
  - For high developed NWS not very important. (fusion enhanced)
  - Possibly opens the gate to gun design
- Compressibility of Uranium lower than Plutonium (EOS + Phase Transition) higher sophisticated implosion design

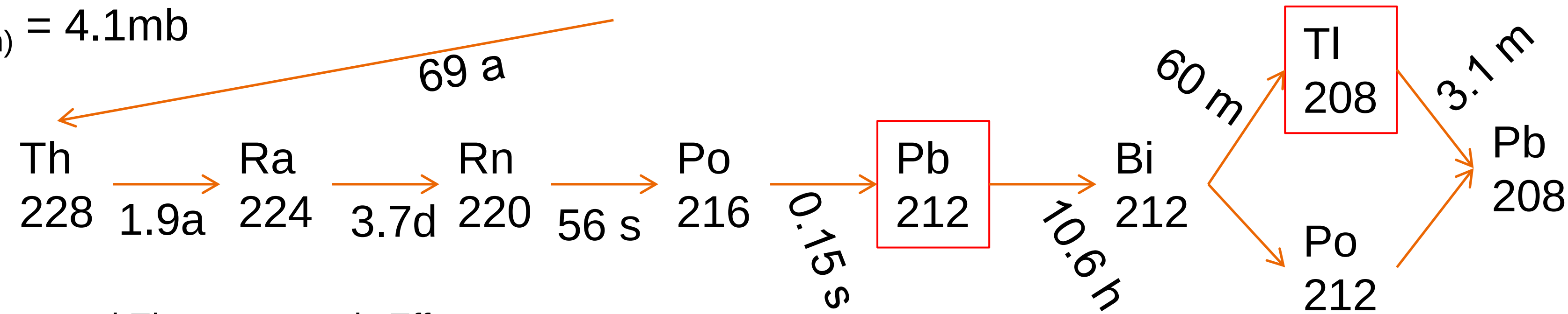
- Urannium-233 is a potent and proven material for nuclear weapon design. It might be used even for low tech gun-design

Good News: U-233 is always contaminated with U-232

U-232 Contamination via (n,2n) reactions



- threshold energy of (n,2n) reactions is approx. 6 MeV
- Half life of U-232 is 69 years
- Decay chain contains strong gamma emitters Pb-212 and Tl-208 (Tl-208: 2614 keV)



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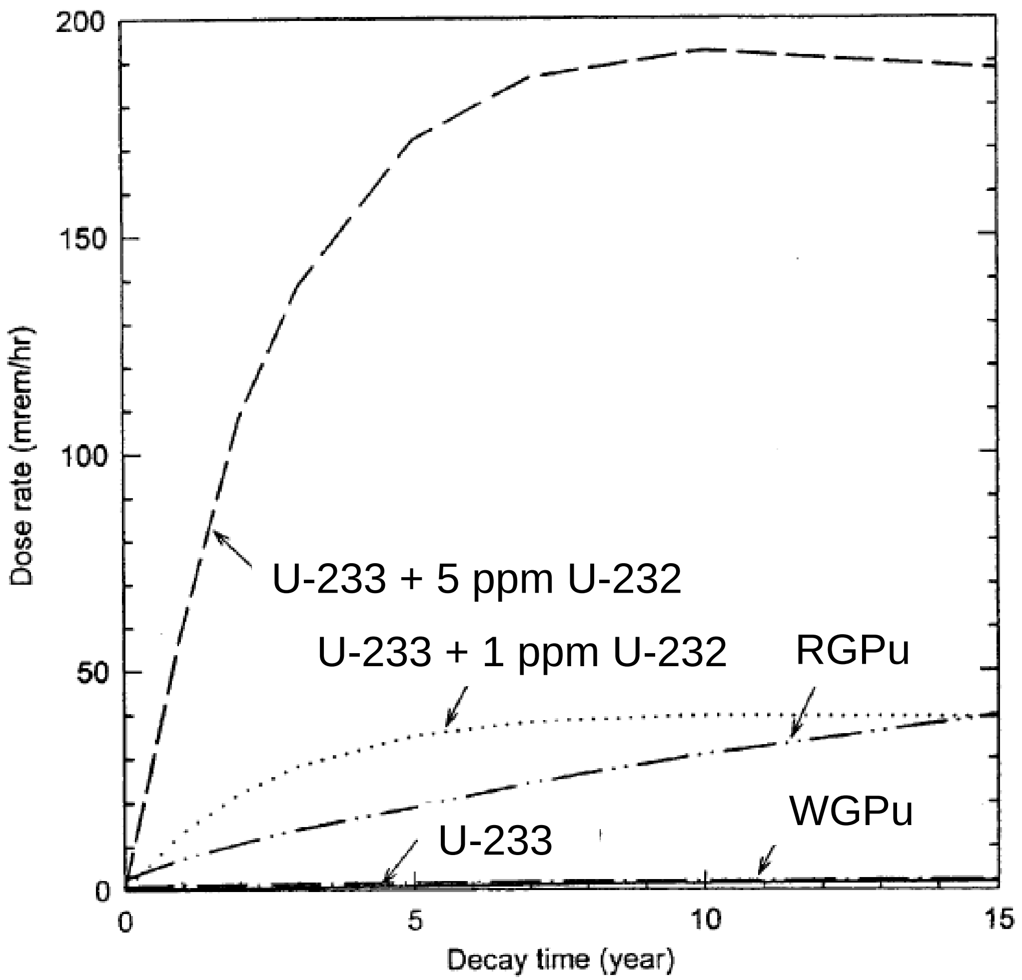
Dose rate as function of cooling time



- Dose rate of 5 kg sphere in 0.5 m distance
- Dose rate mainly from Tl-208 2.6 MeV gammas
- Shielding difficult
  - Reduction to ½ by lead 1.2 cm @ 2.6 MeV
- IAEA Standard for „reduced physical-protection Requirements" of 1 Sv/h @ 1m obtained not until **2.4 %** U-232 (INFCIRC/225/Rev.4)
- Protection in place for short time (5 till 50 years)

Quelle: Kang und von Hippel: U-232 and the Proliferation-Resistance of U-233 in Spent Fuel

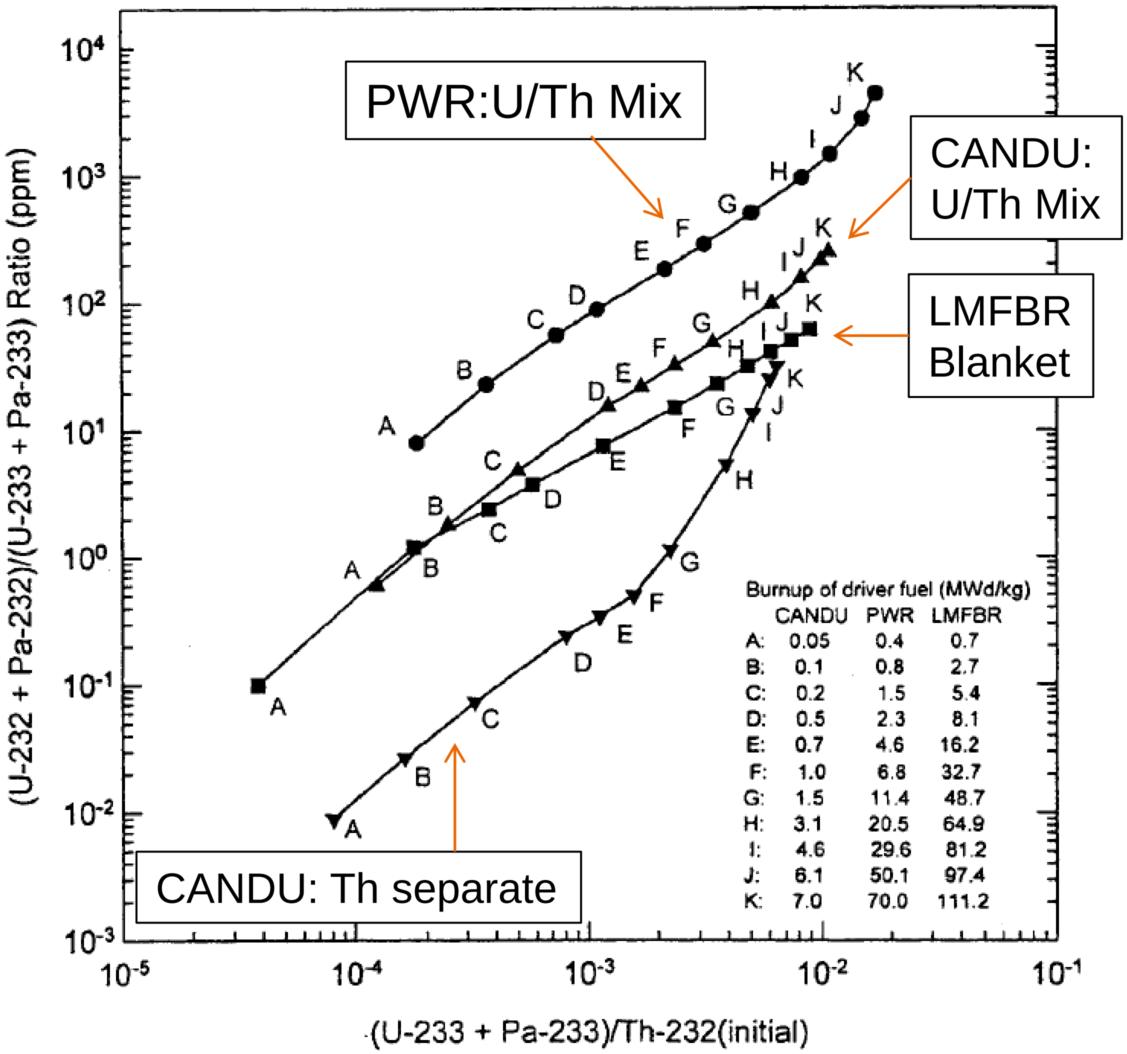
Permitted working hours on core handling (under aspects of radiation protection)

Material	Dose rate	Working hours
WGPu	13 µSv/h	1500 h
RGPu	82 µSv/h	240 h
U-233 + 1 ppm U-232	130 µSv/h	150 h
U-233 + 5 ppm U-232	590 µSv/h	32 h
U-233 + 100 ppm U-232	12.7 mSv/h	90 min
U-233 + 1% U-232	1.3 Sv/h	1 min

5 kg sphere in 0.5 m distance after 1 year cooling time. Permitted maximum dose 20 mSv/year (for working hours)

Quelle: Kang und von Hippel: U-232 and the Proliferation-Resistance of U-233 in Spent Fuel

U-232 / U233 Production



	CANDU	PWR	LMFBR
A:	0.05	0.4	0.7
B:	0.1	0.8	2.7
C:	0.2	1.5	5.4
D:	0.5	2.3	8.1
E:	0.7	4.6	16.2
F:	1.0	6.8	32.7
G:	1.5	11.4	48.7
H:	3.1	20.5	64.9
I:	4.6	29.6	81.2
J:	6.1	50.1	97.4
K:	7.0	70.0	111.2

- U-232 build-up governed by high energy tail of neutron spectrum
- U-233 breeding in Th-232 leads inevitable to fissile material in Thorium and thus U-232 build-up
- Well moderated reactors or blanket concepts reduce the U-232 content

Quelle: Kang und von Hippel: U-232 and the Proliferation-Resistance of U-233 in Spent Fuel

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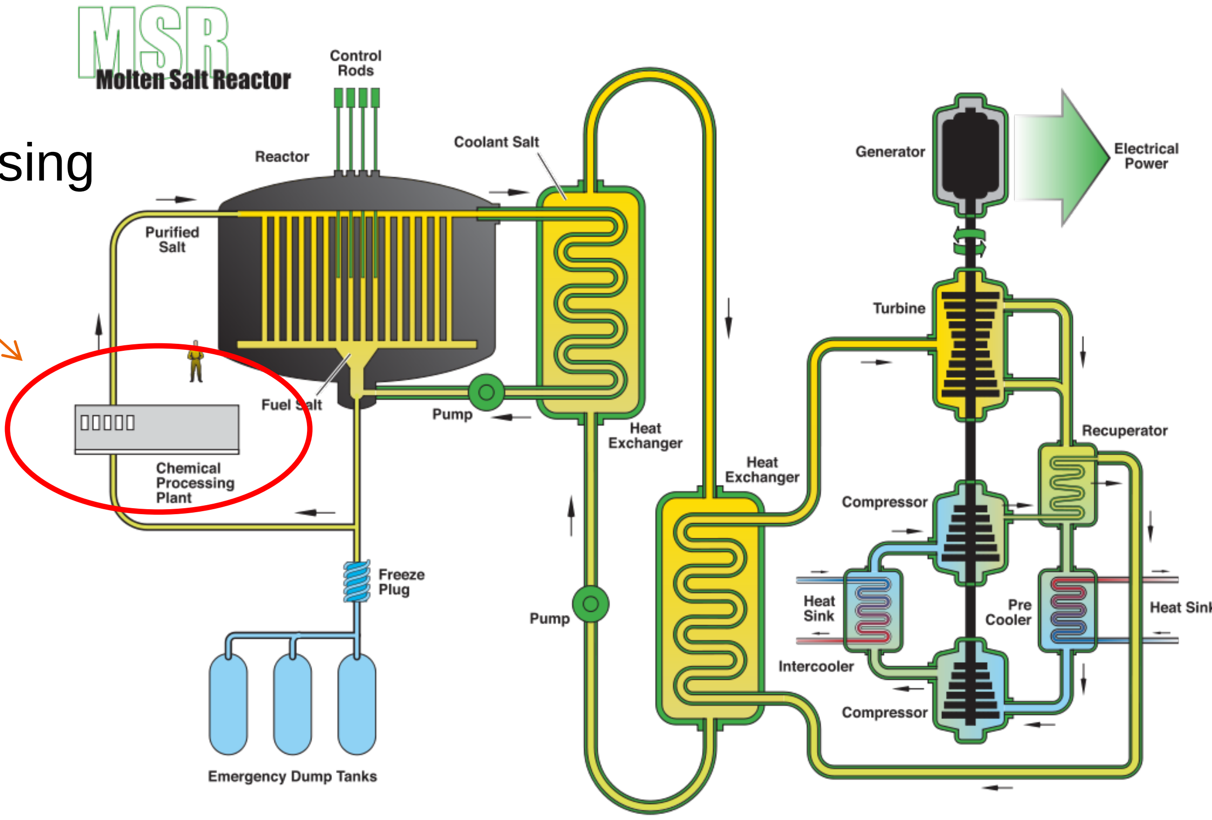
Special case: Online Reprocessing

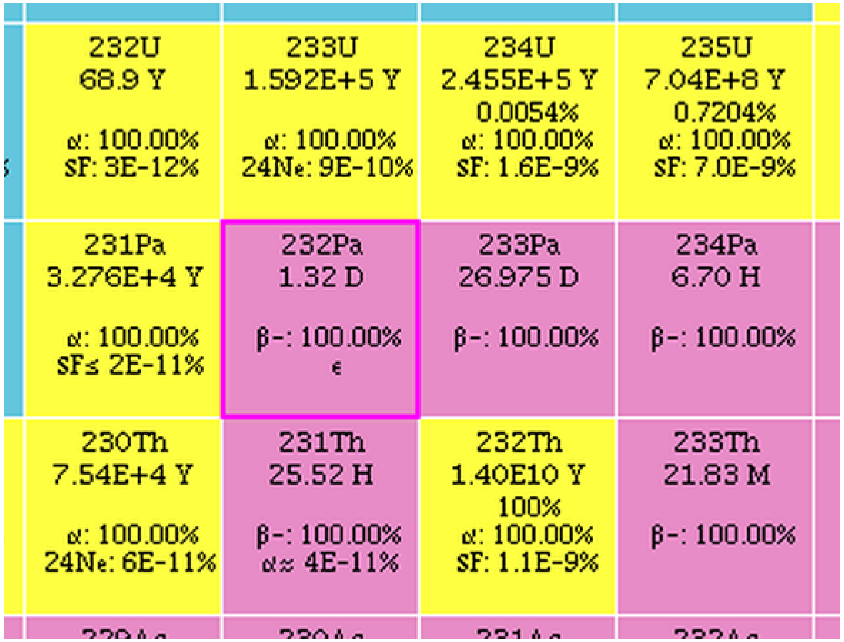
Online Chemical Reprocessing

- One Alternative is 3 Stream Process: Th, Pa, and U separated
- Pa is Pa-231, Pa-232, Pa-232



Generation IV: Molten Salt Reactor

Special case: Online Reprocessing



232U 68.9 Y α: 100.00% SF: 3E-12%	233U 1.592E+5 Y α: 100.00% 24Ne: 9E-10%	234U 2.455E+5 Y 0.0054% α: 100.00% SF: 1.6E-9%	235U 7.04E+8 Y 0.7204% α: 100.00% SF: 7.0E-9%
231Pa 3.276E+4 Y α: 100.00% SF≤ 2E-11%	232Pa 1.32 D β-: 100.00% ε	233Pa 26.975 D β-: 100.00%	234Pa 6.70 H β-: 100.00%
230Th 7.54E+4 Y α: 100.00% 24Ne: 6E-11%	231Th 25.52 H β-: 100.00% α≈ 4E-11%	232Th 1.40E10 Y 100% α: 100.00% SF: 1.1E-9%	233Th 21.83 M β-: 100.00%

- Pa-231 decays by alpha
- Pa-232 decays by β- to U-232 in 1.32 days
- Pa-233 decays by β- to U-233 in 27 days

Back of Envelope calculation

Initial solution 2% Pa-232

- Let Protactinium solution cool for 33.8 days
- Separate Uranium (high U232 Content)
- Let Solution cool for 1 year
- Separate Uranium (ultra low U-232)

Fraction 1: 3.4 % U-232
Fraction 2: 0.1 ppm U-232
42 % of U-233 recovered

Quelle: http://www.nndc.bnl.gov Nudat2

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Further difficulties

- Dilution with U-238, resulting Uranium is “LEU”
  - Generation of Plutonium and further minor actinides
  - Reduction of breeding efficiency
  - Some schemes call for 19.5 % LEU as initial fuel

- Permanent disposal site transforms after 700 years to superior U-233 mine
  - Half life of U-232 to short for effective long term protection

- Large fraction of U-232 undesirable for industry in first place.
  - Large gamma activity call for remote handling of used and reprocessed fuel elements → higher costs
  - Economic vs. Proliferation

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Conclusions

- Renewed interest in Thorium cycle
- Fissile Uranium-233 is breed from Thorium-232
- Uranium 233 is a potent nuclear weapon material
- Uranium-232 is produced in conjunction with U-233
- Handling difficulties due to gamma emitting Uranium-232
- But U-232 is not a very effective safeguarding agent

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Final

Thank You very much for Your interest.

We will be happy to answer Your questions.

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