JOLIOT CURIE SCHOOL – September 2019

NUCLEAR FUEL CYCLE(S)

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NUCLEAR FUEL CYCLE : *MAIN STAKES*

Nuclear fuel cycle:

- *feeding nuclear reactors (the « front end »)*
- *managing used fuels (the « back end »)*

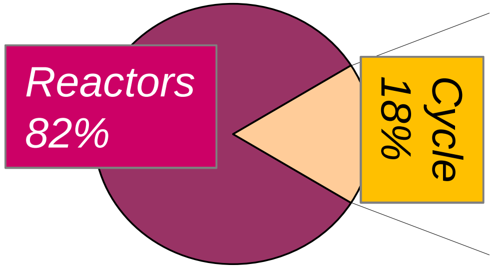
- an essential issue for *sustainability requirements*

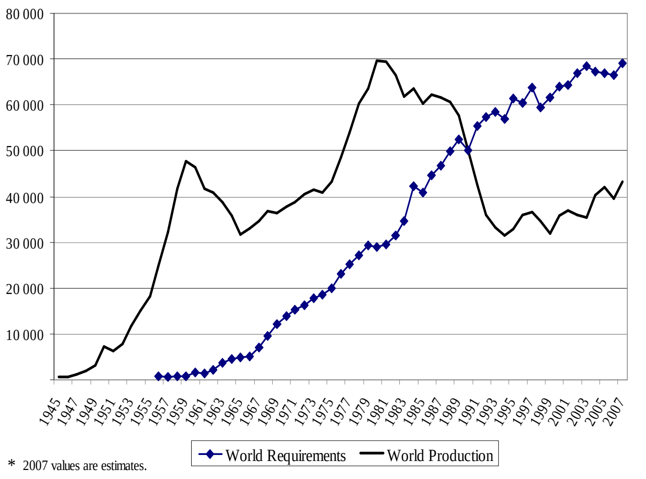
- *natural resource preservation*

- *environmental impact minimization*

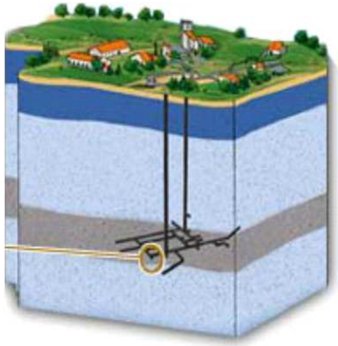
- *proliferation risks minimization*

an evaluation
of french electricity production (levelized) cost
(CEA, 2012 report to French Gov.)





* 2007 values are estimates.



NUCLEAR FUEL CYCLE

1 – GENERAL INTRODUCTION

(about nuclear energy, about nuclear fuels…)

2 – FUEL CYCLE FRONT-END

(from mining to fuel fabrication…)

3 – USED NUCLEAR FUEL

(in-pile fuel changes…)

4 – USED NUCLEAR FUEL MANAGEMENT

(fuel cycle options, reprocessing and recycling French option)

5 – PARTITIONING & TRANSMUTATION

(long-lived nuclides dedicated management: what for, and how ?)

6 - SEPARATION , HOW ?

(an introduction to actinide separation processes)

DE LA RECHERCHE À L'INDUSTRIE

NUCLEAR FUEL CYCLES

1 – GENERAL INTRODUCTION

(about nuclear energy and nuclear fuels)

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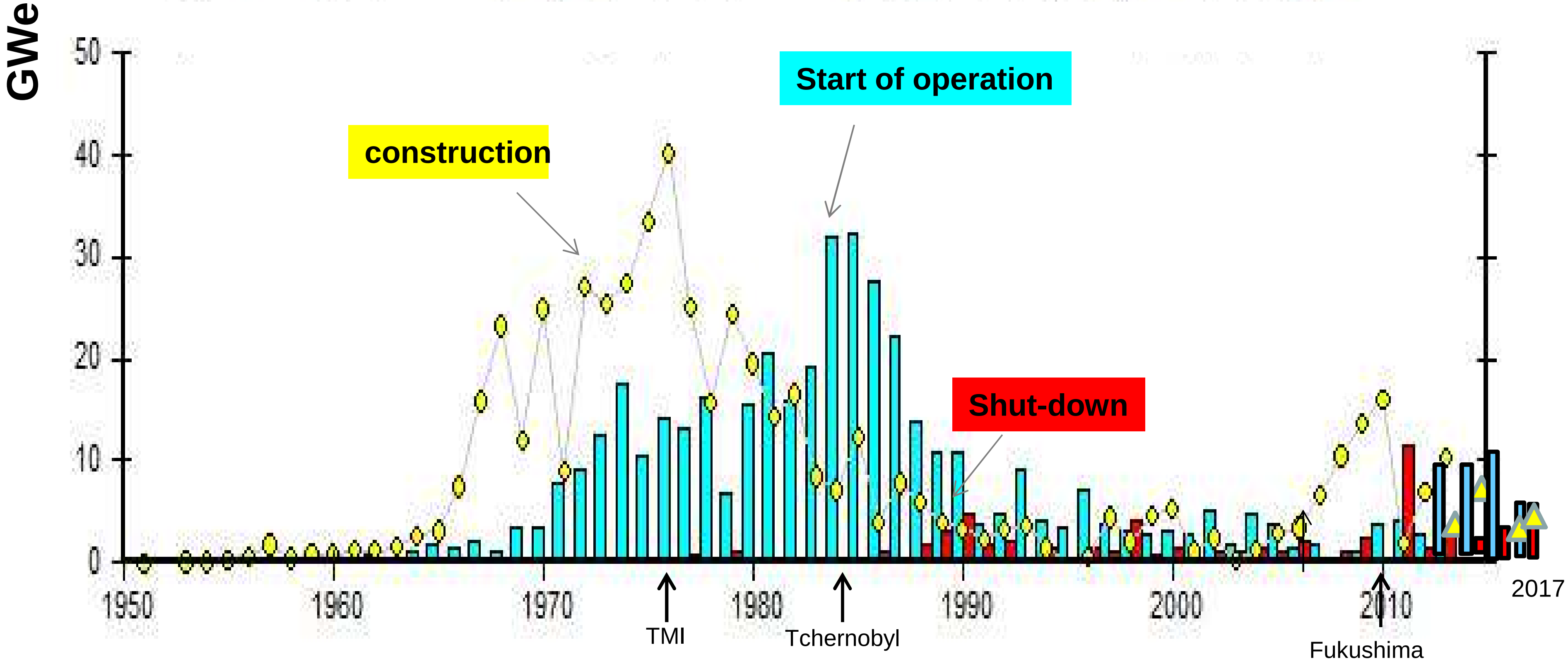
Joliot Curie School, St Pierre d'Oleron, 22-27 September, 2019

A « HISTORY » OF NUCLEAR ENERGY…

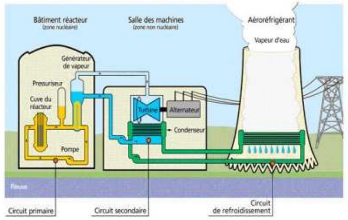


Generation I

(dismantling)

→

Generation II



(operation)

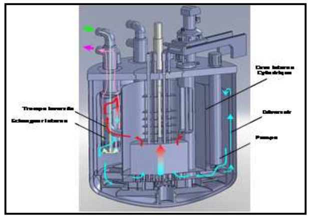
→

Generation III

(construction)

→

Generation IV

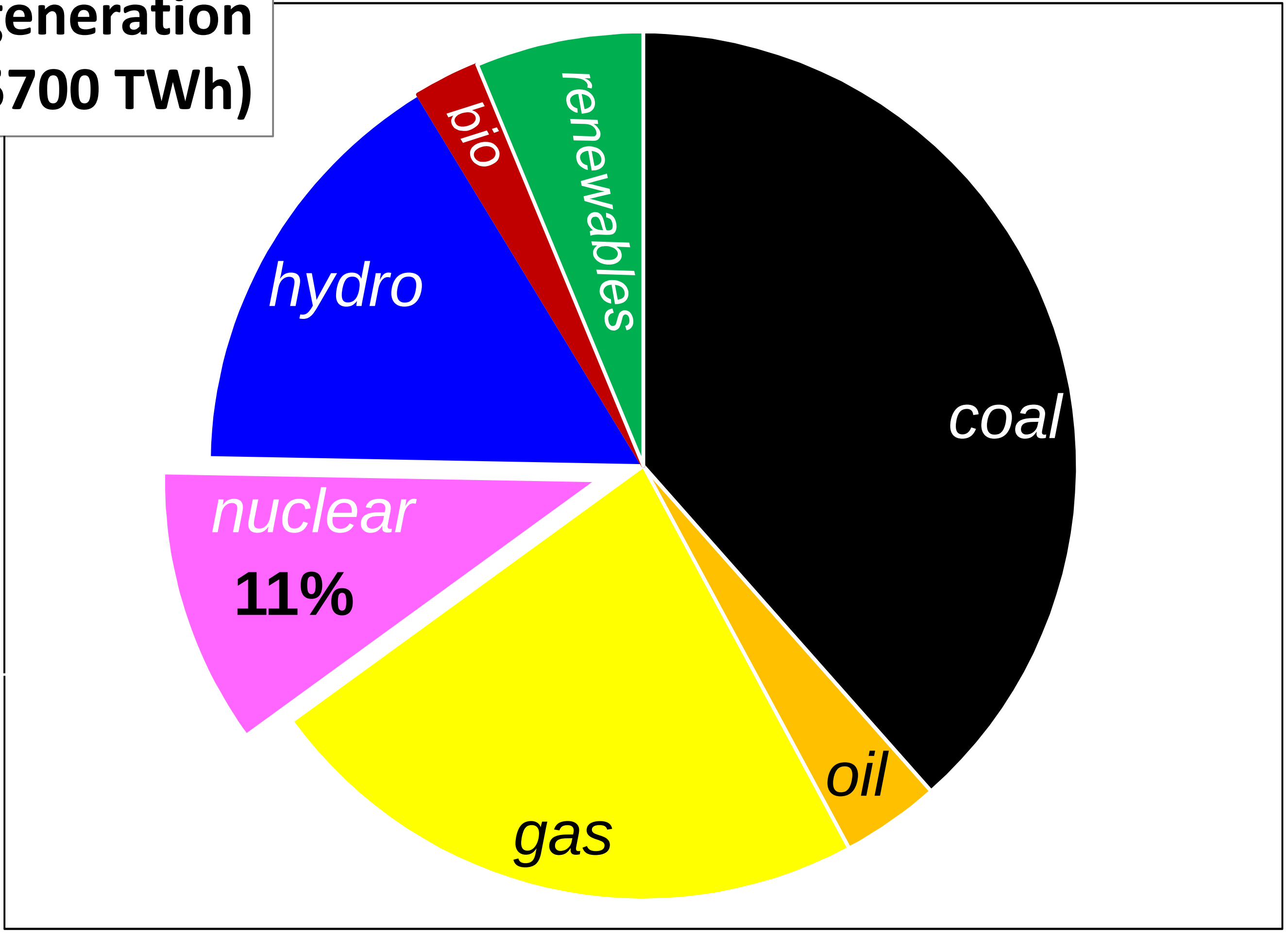


(studies and prospects)

NUCLEAR ENERGY TODAY

IEA, World Energy Outlook, 2018 (2017 estimate)

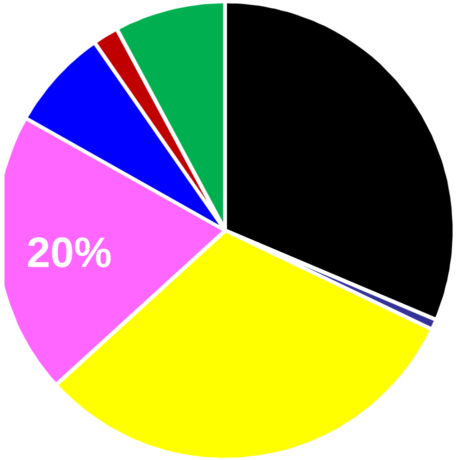
Power generation (25700 TWh)



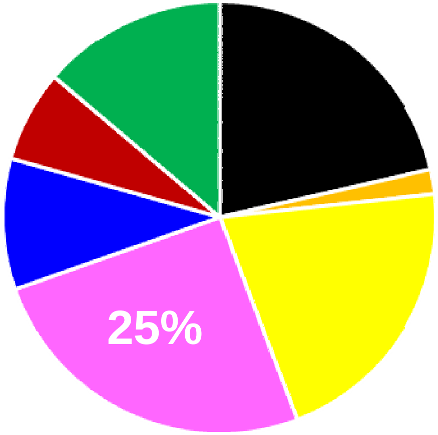
POWER GENERATION IN DIVERSE COUNTRIES

IEA, World Energy Outlook, 2018 (2017 estimate)

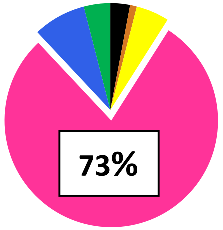
USA (4300 TWh/an)



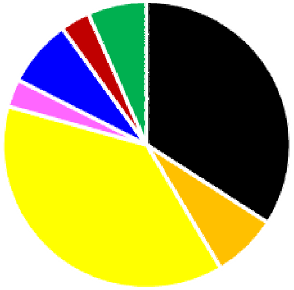
EUROPEAN UNION(3300)

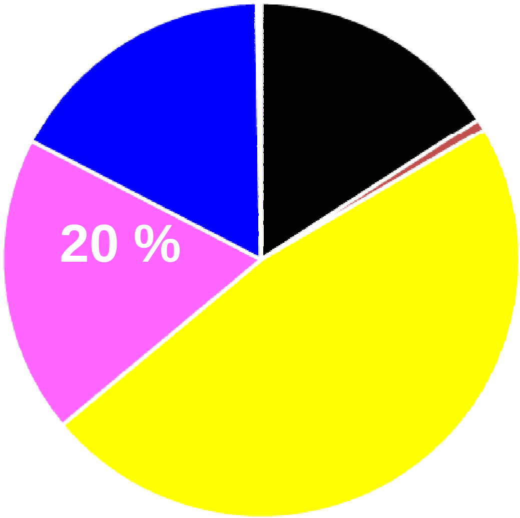


FRANCE (530)

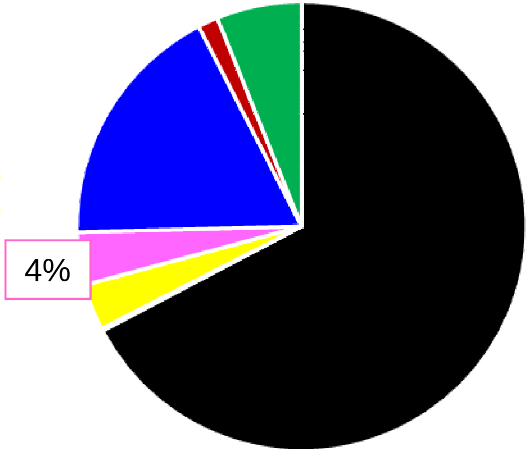


JAPAN (1100)

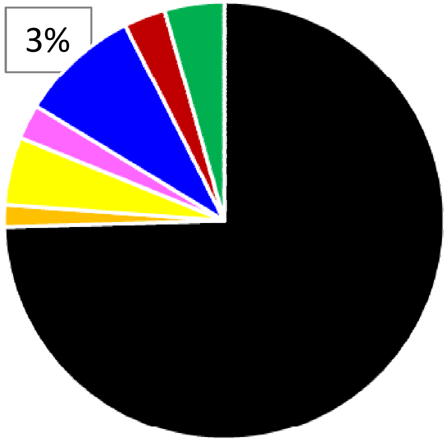




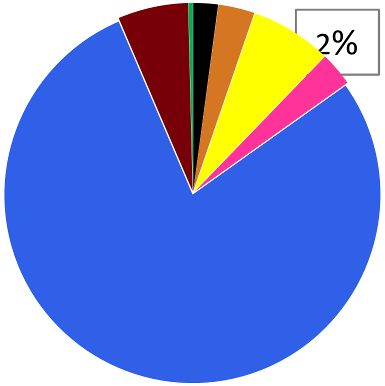
RUSSIA (1100)



CHINA (6600)



INDIA (1600)



BRAZIL (590)

NUCLEAR FUELS : *REQUIREMENTS*

(1) a **fissile** nucleus : ^{235}U available *(^{239}Pu , ^{233}U)*

Nucléide	σa (barns)	σf (barns)
^{232}Th	7.40	0
^{231}Pa	227	0.01
^{233}Pa	41.5	0
^{232}U	149.7	77.1
^{233}U	571.1	525.2
^{234}U	103.5	0.46
^{235}U	681.5	582.6
^{236}U	5.21	0.047
^{238}U	2.719	0.000012
^{237}Np	181.0	0.018
^{238}Pu	563.4	17.3
^{239}Pu	1017.7	747.3
^{240}Pu	288.8	0.068
^{241}Pu	1375.3	1012.3

(neutron velocity 2km/s)

RADIONUCLEDES	PERIOD (years)
^{242}Pu	3,74 10^{5}
^{241}Pu	14,4
^{240}Pu	6,56 10^{3}
^{239}Pu	2,41 10^{4}
^{238}Pu	87,7
^{237}Np	2,14 10^{6}
^{238}U	4,47 10^{9}
^{236}U	2,34 10^{7}
^{235}U	7,04 10^{8}
^{234}U	2,46 10^{5}
^{233}U	1,59 10^{5}
^{232}U	70

NUCLEAR FUELS : *REQUIREMENTS*

(1) a **fissile** nucleus : ^{235}U available *(^{239}Pu , ^{233}U)*

(2) … in a particular compound

- high **density of fissile nuclei** (purity, enrichment)
- avoiding presence of **neutron poisons** (O, N, C suitable)
- able to provide efficient **heat transfer** (conduction)
- **no chemical reactions** with environment (chemically "inert")
- **stable under irradiation** conditions (n, αβγ, and Θ)
- … particular requirements – passive safety- (Doppler effect…)

Nuclide	σa (barns)
Natural hydrogen	0.332
Deuterium	0.000506
Boron 10	3840
Natural carbon	0.00337
Natural oxygen	0.000191
Natural zirconium	0.184

(neutron velocity 2km/s)

NUCLEAR FUELS : *WHICH COMPOUND ?*

(1) at the beginning : **metallic** natural uranium fuels
(France: gas cooled reactors (UNGG) ,…)

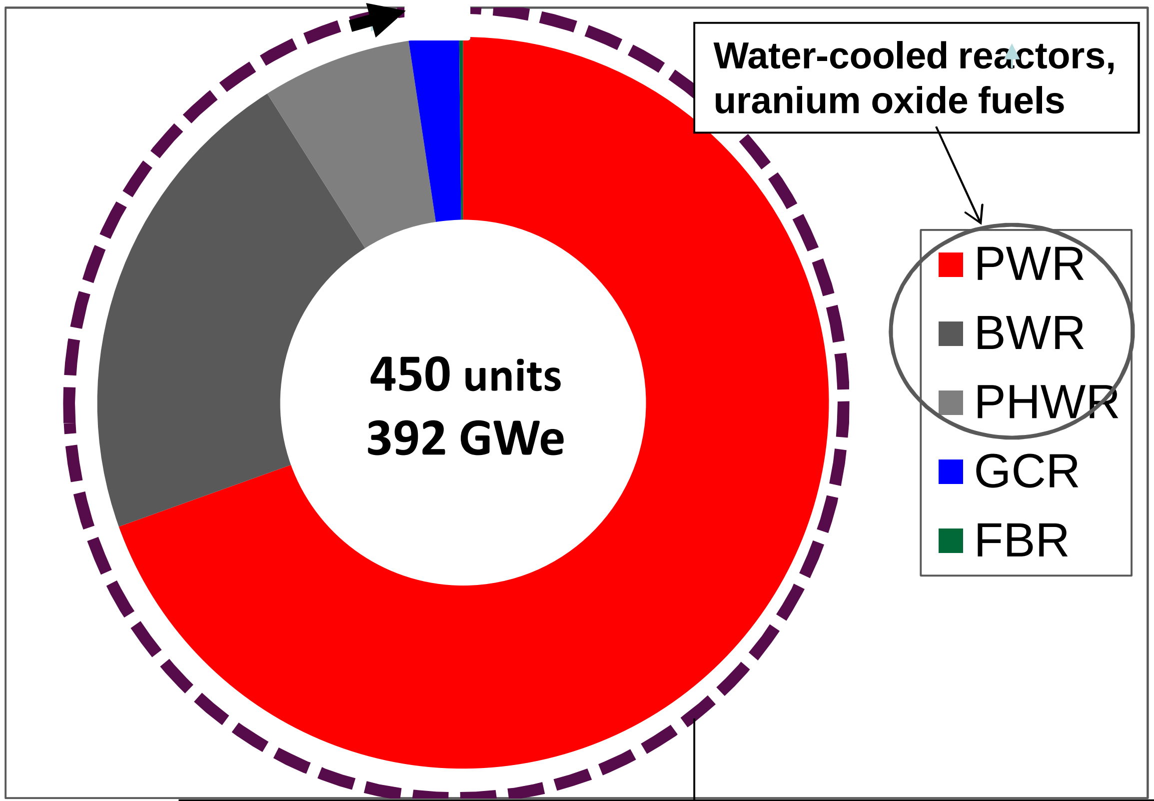
(2) mainly operated today : uranium (natural or enriched) **oxide** fuels
(Heavy water reactors (CANDU), Light water reactors (P or B))

(3) Other possible fuels : plutonium fuels (MOX), ^{233}U fuels (INDIA)
other ceramic fuels (carbide, nitride,…)?
liquid fuels (MSR :fluoride, chloride,…)

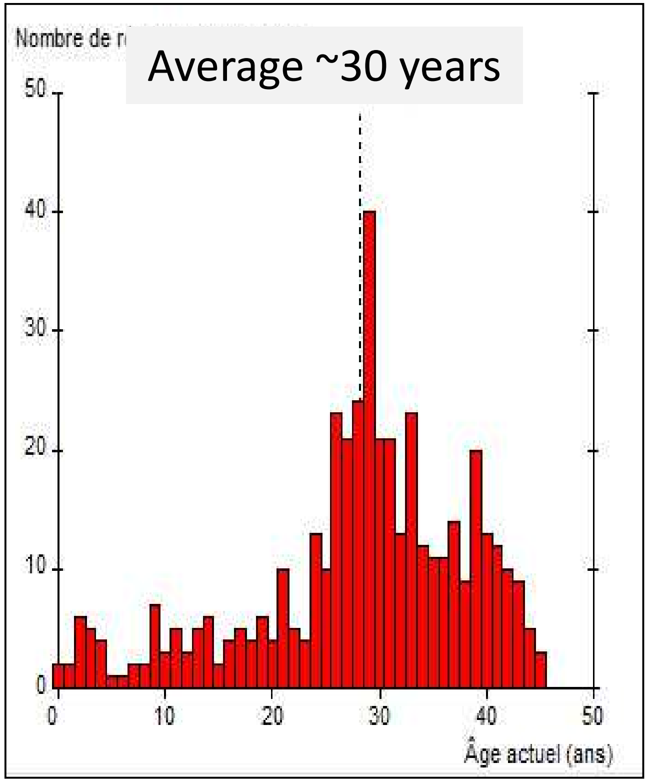
Nature of the fuel	UO_2	UC	UN	U
U concentration (g/cm^3)	9,7	12,9	13,5	19
Fusion temperature(°C)	2 775	2 480	2 780	1 160
Thermal conductivity at 1000°C(W.m^{-1}.K^{-1})	2,9	19,6	19,8	30
Thermal expansion (from 20°C to 1000°C) (10^{-6} / °C)	12,6	12,4	10,0	16,5
Compatibilité avec l'eau	+	-	-	- -

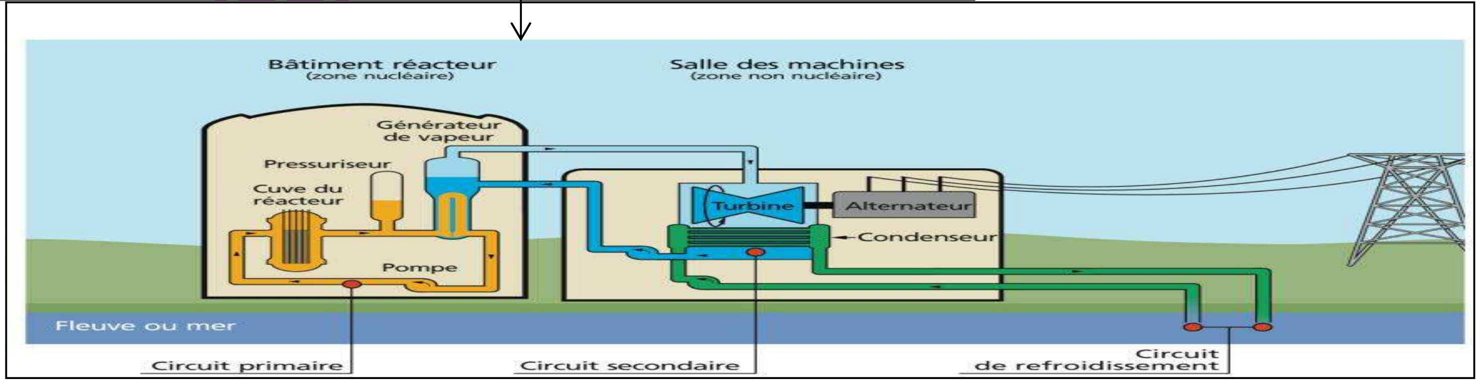
(some important diverse fuel properties)

WORLD NUCLEAR FLEET…

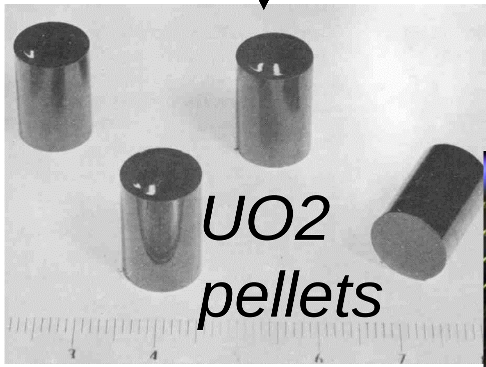


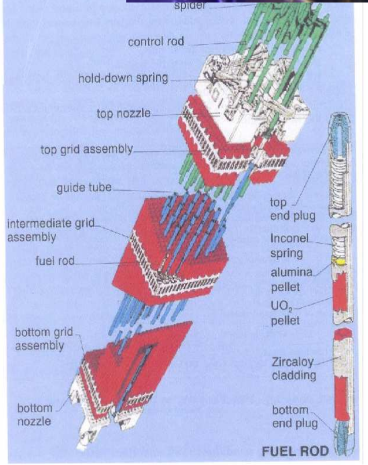
Water-cooled reactors, uranium oxide fuels


Average ~30 years

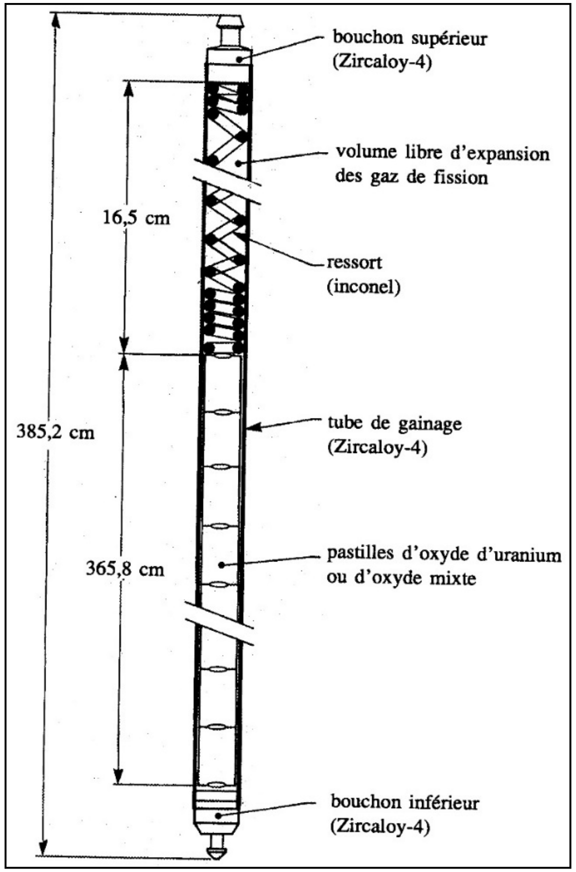


PWR UO2 FUEL





PWR
sub-assembly



PWR 900
fuel rod scheme

PWR (900 MWe) : *some FUEL FEATURES*

- **coolant :** **water** ***(~155 bar ; ~300°C)***
- **Moderator :** **water**

- **FUEL :** **UO_2** ***(^{235}U ~4 %)***

- **CLADDING :** **zircaloy *(Zr ; Sn, Fe, Cr, …)***

- **Sub-assemblies:** **square 17 x 17 ; 264 rods**
  rod height: 3.8 m to 4.5 m
  rod diameter: ∅ext ~ 9.5 mm, thickness : 0.6 mm
  gas expansion chamber : pressurized 25 bars

- **Pellets :** **∅ 8 mm h ~ 13.5 mm**
  ρ = 10.9 g/cm^3
  residual porosity ~ 5 %
  extracted power *several 100 W/cm^2*

- **Reactor** **157 sub-assemblies *(> 40 000 rods)***

URANIUM ANNUAL NEEDS

- WORLD FLEET
  - fissions per year
  - ^{235}*U consumption*
  - *fission products amount*

448 units, 392GWe, 2488TWhe

- FRENCH FLEET

58 units, 63GWe, 380TWhe

URANIUM ANNUAL NEEDS

- WORLD FLEET
  - fissions per year
  - *^{235}U consumption*
  - *fission products amount*

(~6.6 times French data:
FP~330 tons)

448 units, 392GWe, 2488TWhe

- FRENCH FLEET
- fissions :1.3 10 ^{29}
  (conversion yield+33%)
  (1 fission: 200 MeV)
- Fissile ^{235}U consumption :50 tons
- FP amount: 50 tons

58 units, 63GWe, 380TWhe

DE LA RECHERCHE À L'INDUSTRIE

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NUCLEAR FUEL CYCLES

2 –FRONT-END OPERATIONS
(from mining to fuel fabrication)

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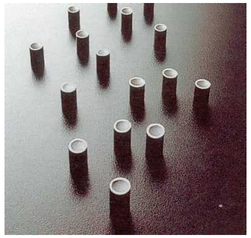
FRONT-END : *AN OVERVIEW*



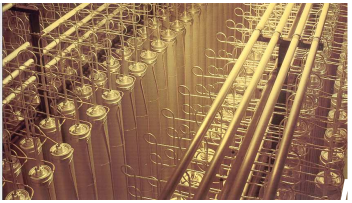
REACTORS

UOX# 10 000 t per year

(french fleet: # 1000 t per year)

Fabrication

^{235}U# 4%



Uranium enrichment

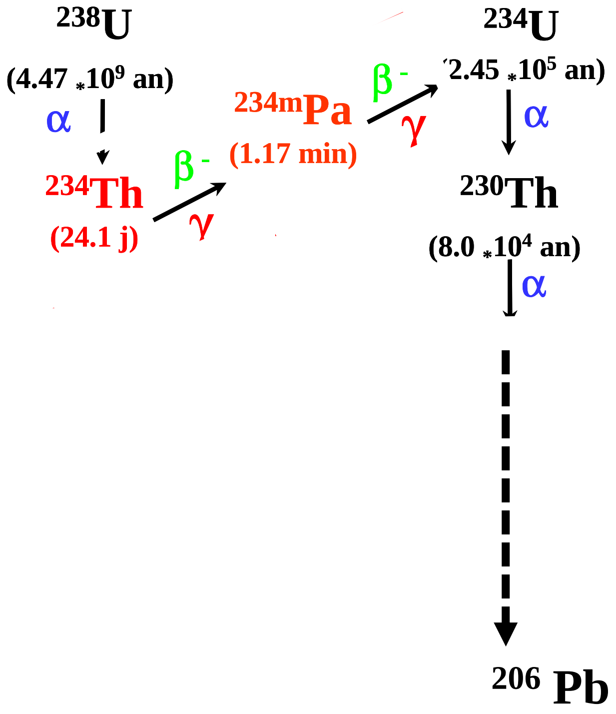
^{235}U= 0.7%

Mining and refining

- <u>many transformation steps</u>, from ore to fuel
- *a competition market*
- *important improvement stakes*

URANIUM ISOTOPS

ISOTOP	HALF-LIFE (years)
^{238}U	$4,47\ 10^{9}$
^{236}U	$2,34\ 10^{7}$
^{235}U	$7,04\ 10^{8}$
^{234}U	$2,46\ 10^{5}$
^{233}U	$1,59\ 10^{5}$
^{232}U	70



NATURAL **URANIUM ISOTOPS :**

^{238}U (99.28%) ^{235}U (0.71%) *^{234}U (0.054%)*

NATURAL URANIUM :
GEOLOGY / GEOCHEM ISTRY

NATURAL (PRIMORDIAL) URANIUM : ***MAINLY IN EARTH CRUST***
(chemical affinity → silica)

PRIMARY ORE : CRYSTALIZATION *(granitic rocks)*

URANIUM « HISTORY »:

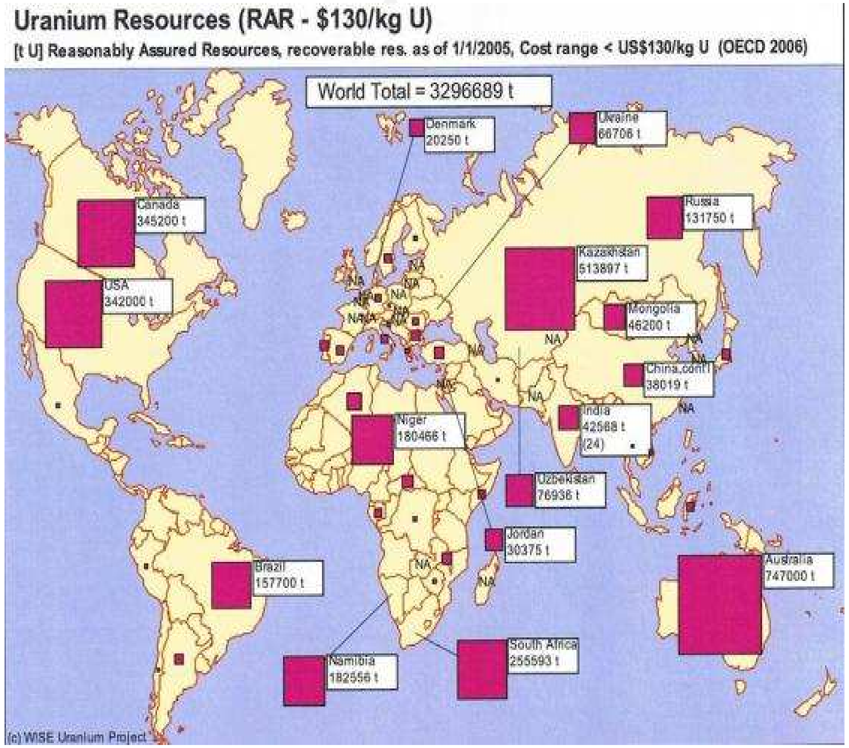
- **Oxidized compounds: UO_2 and UO_3 (U3O8)**
- **UO2 hardly soluble (U4+)**
- **UO_3 soluble (ion UO_2^{2+}) and mobile *(hydrothermal fluids)***
- **Secondary ore(s): sedimentary *(mined from about 0.1%)***

Particular case: conjunction (abundant hydro-transport/reductant)
UO2 (not soluble)precipitation
possibly very high U concentrations (>20%)

URANIUM RESOURCE

Extraction cost (US $ / Kg U)	Conventionnal resources (Mt)	
	Identified resources	**Undiscovered resources**
< 40	0,7	
40 – 80	1,3	
80 - 130	4	
130 -260	1,7	
TOTAL	7,6	7,7
	15,3	

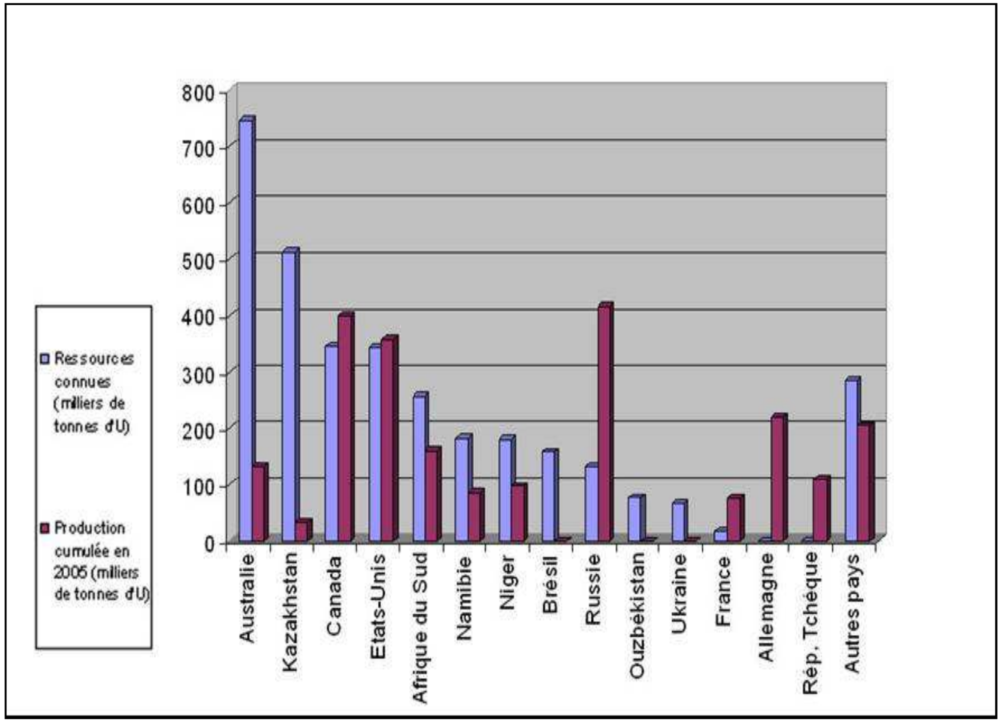
AIEA, « red book », 2014



Uranium Resources (RAR - $130/kg U)

[t U] Reasonably Assured Resources, recoverable res. as of 1/1/2005, Cost range < US$130/kg U (OECD 2006)

(c) WISE Uranium Project



URANIUM RESOURCE

Current annual needs:~70kt

and non conventional resources : phosphates (→8 M t ?)

sea water ? :4 Gt (but 3.3µg/l)

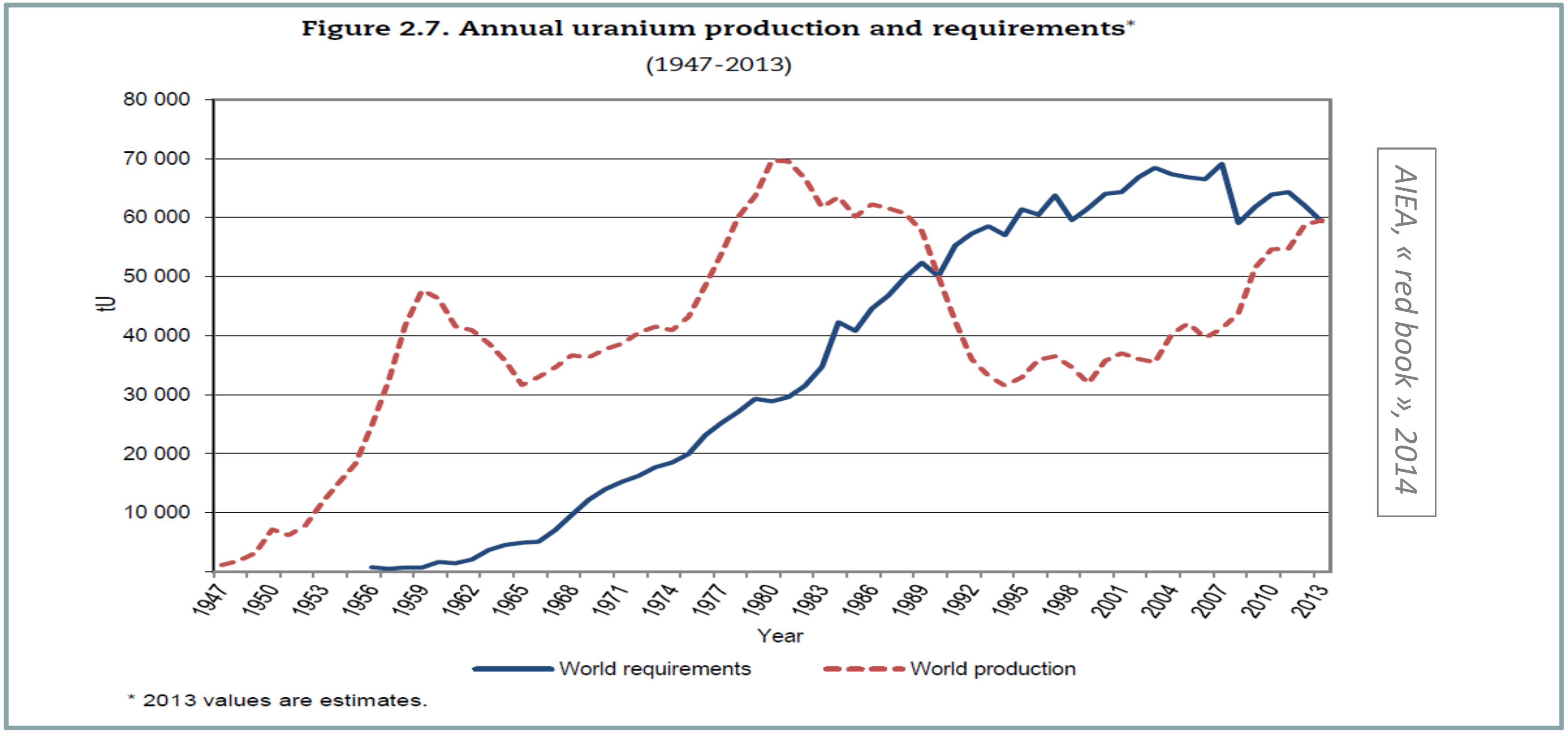


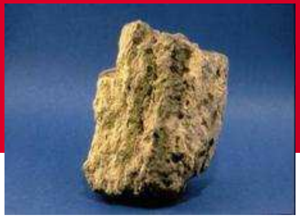
Figure 2.7. Annual uranium production and requirements*

(1947-2013)

* 2013 values are estimates.

AIEA, « red book », 2014

MINING OPERATIONS



(#60%)

CRUSHING

GRINDING

DYNAMIC LEACHING

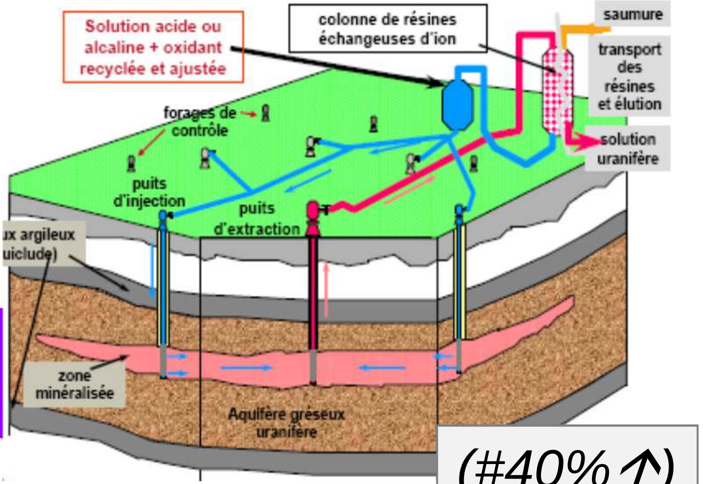
S/L SEPARATION

CRUSHING

LEACHING

S/L SEPARATION

IN SITU RECOVERY



(#40% ↑)

Champ de forage ISR

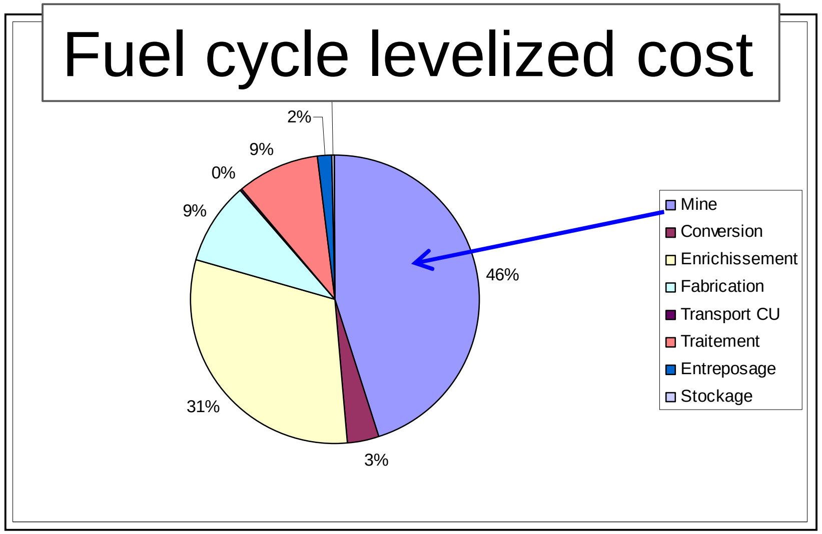
REFINING (EXTRACTION/BACK-EXT)

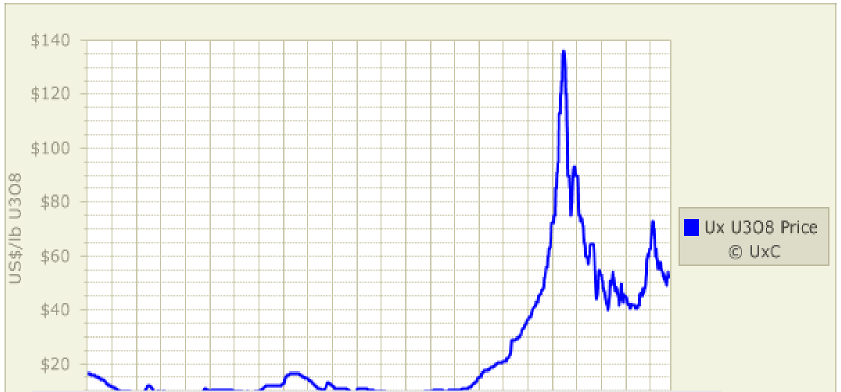
PRECIPITATION

to get 1000 t U /year…

Mining ore 15%	18tons ore/day
Mining ore 0.4%	685 tons ore/day
Mining ore 400ppm	6850 tons ore/day
ISL	56000 m3/day

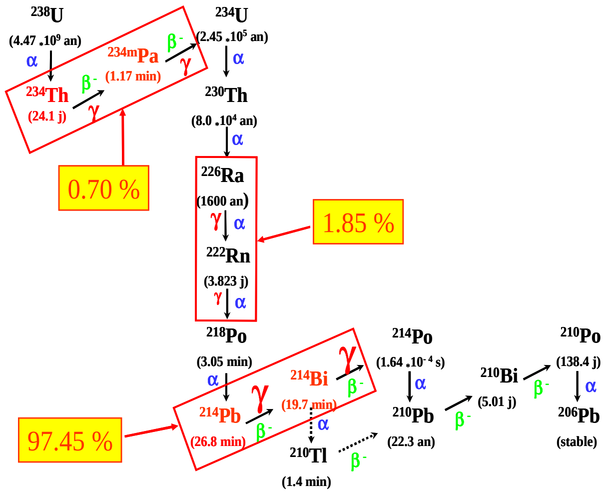
MINING OPERATIONS

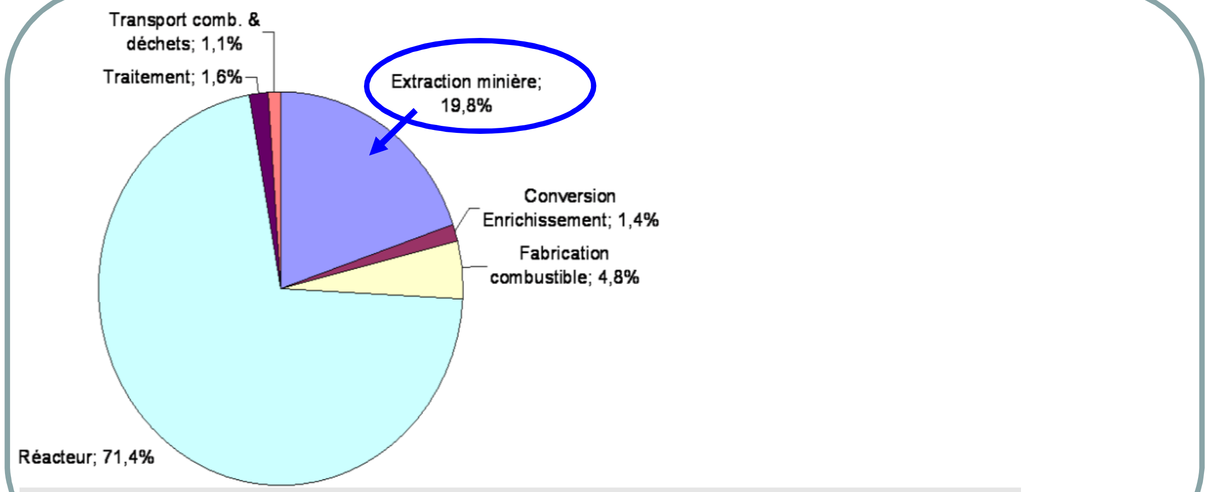

Fuel cycle levelized cost



Uranium « spot » price

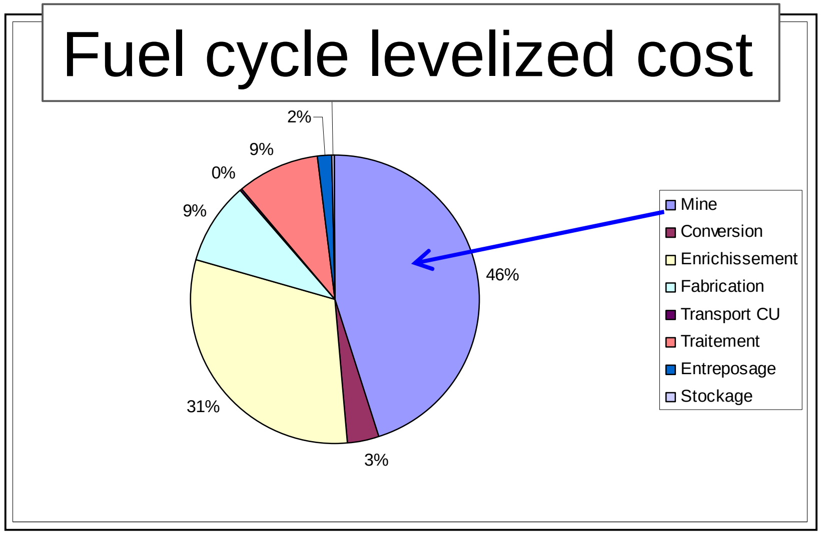
Mining in nuclear energy radiologic status





French fleet collective dose, 2009 **(0,14h.Sv/TWh)**

MINING OPERATIONS



Fuel cycle levelized cost

Uranium « spot » price

Levelization: if 5%/year

Year 1 (current year) : cost = C

Year 2: cost = C/1.05

Year n : cost = $C/(1.05)^n$

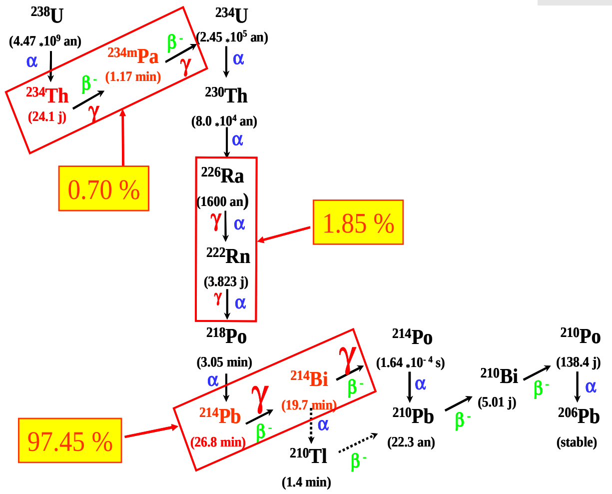
Year 50:cost= $C/(1.05)^{50} = C/ 10$

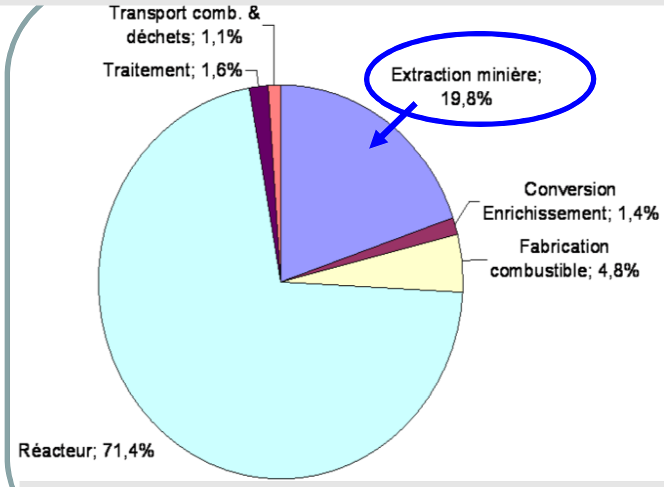
Utilities:

long term supply agreements

MINING OPERATIONS

Mining in nuclear energy radiologic status





French fleet collective dose, 2009 **(0,14h.Sv/TWh)**

power generation: 400 TWh
N workers involved
Σ worker doses: 56 Sv
“collective dose”:
56/400 = 0.14 Sv per TWh

MINING : *RESEARCH DRIVERS*

- ***a general statement*** *: a rather “old” industry*
  (technologies, environmental issues)

- ***but currently depressed market*** *: overcapacities,*
  no possibilities for large investments

- ***some identified research goals*** *:*
  - *exploration (notably for deep mining)*
  - *decrease water consumption (on-site yellow-cake elaboration)*
  - *Uranium recovery as other mines by-products (phosphates)*
  - *Long term goal: Uranium recovery from seawater !!*

MAIN CHEMICAL REACTIONS INVOLVED *(1/4)*

1. URANIUM ORE LEACHING

ALKALI LEACHING *(NaHCO3+Na2CO3)*

$UO_3 + Na_2CO_3 + 2\ Na\ HCO_3 \rightarrow UO_2\ Na_4(CO_3)_3 + H_2O$

$UO_2 + ½\ O_2 + Na_2CO_3 + 2\ Na\ HCO_3 \rightarrow UO_2\ Na_4(CO_3)_3 + H_2O$

ACID LEACHING *(H_2SO_4)*

$UO_3 + H_2SO_4 \quad H_2SO_4 \quad \rightarrow \quad UO_2\ SO_4 + H_2O$

$UO_2 + Fe_2\ (SO_4)_3 \rightarrow UO_2\ SO_4 + 2\ Fe\ SO_4$

$6\ Fe\ SO_4 + 3\ H_2SO_4 + NaClO_3 \rightarrow 3\ Fe_2(SO_4) + NaCl + 3\ H_2O$

MAIN CHEMICAL REACTIONS INVOLVED *(2/4)*

2. URANIUM REFINING *(SOLVENT EXTRACTION)*

***EXTRACTION BY TERTIARY AMINES** (R_3N)*

$$UO_3\,Na_4(CO_3)_3 + 3\,H_2SO_4 \rightarrow UO_2\,SO_4 + 2\,Na_2SO_4 + 3\,CO_2 + 3\,H_2O$$

species in aqueous solution : $UO_2\,SO_4$ $[UO_2\,(SO_4)_2]^{2-}$ $[UO_2\,(SO_4)_3]^{4-}$

$2\,\overline{R_3N} + H_2SO_4 \rightarrow \overline{(R_3NH)_2SO_4}$ *[acidification of ternary amine]*

$2\,\overline{(R_3NH)_2SO_4} + [UO_2(SO_4)_3]^{4-} \rightarrow \overline{(R3NH)_4UO_2(SO_4)_3} + 2SO_4^{2-}$ *[extraction by substitution*

$2\,\overline{(R_3NH)_2SO_4} + [UO_2(SO_4)_3]^{2-} \rightarrow \overline{(R3NH)_4UO_2(SO_4)_3} + SO_4^{2-}$

***EXTRACTION BY NEUTRAL EXTRACTANT** (TBP)*

$$UO_2\,Na_4(CO_3)_3 + 6\,HNO_3 \rightarrow UO_2\,(NO_3)_2 + 4\,Na\,NO_3 + 3\,CO_2 + 3\,H_2O$$

$$UO_2^{2+} + 2\,NO_3^{-} + 2\,\overline{TBP} \rightarrow \overline{UO_2\,(NO_3)_2\,,\,2TBP}$$

MAIN CHEMICAL REACTIONS INVOLVED *(3/4)*

3. URANIUM PRECIPITATION

diuranate d'ammonium

$2\ UO_2\ (NO_3)_2 + 6\ NH_3 + 3\ H_2O \rightarrow (NH_4)_2\ U_2O_7 + 4\ NH_4\ NO_3$

hydroxyde

$UO_2\ SO_4 + MgO + x\ H_2O \rightarrow UO_3,\ x\ H_2O + Mg\ SO4$

$UO_2\ (NO_3)_2 + 5\ H_2O_2 \rightarrow UO_4,\ 4\ H_2O + 2\ HNO_3 + 2\ O_2$

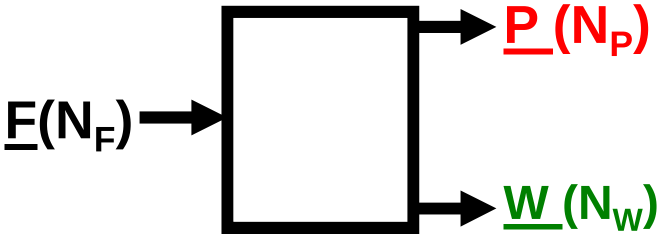
MAIN CHEMICAL REACTIONS INVOLVED *(4/4)*

4. URANIUM CALCINATION

$$(NH_4)_2U_2O_7 \xrightarrow{350^\circ C} 2UO_3 + 2NH_3 + H_2O$$

$$UO_3 \xrightarrow{900^\circ C} U_3O_8$$

URANIUM ENRICHMENT



$\underline{F}(N_F)$

$\underline{P}\ (N_P)$

$\underline{W}\ (N_W)$

P, product (enriched)
W, waste (depleted)
F, feed (natural)

$N_{F,P,W}$: $^{235}U/U$ in F,P,W

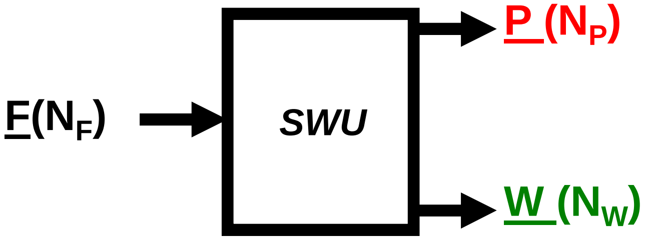
$F = P + W$ *(uranium mass balance)*

$F.N_F = P.N_P + W.N_W$ *(235U mass balance)*

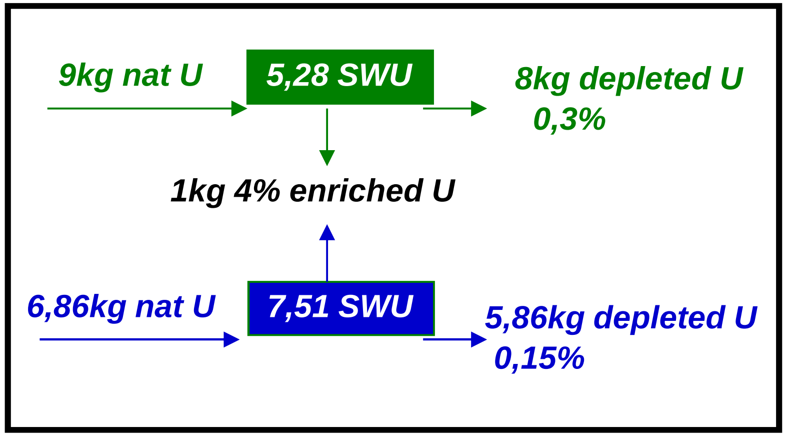
$$\boxed{F = P\ (N_P - N_W)\ /(N_F - N_W)}$$

? *known* *known(4%)* *known(0.7%)* *to be defined*

URANIUM ENRICHMENT



$$F = P\ (N_P - N_W)\ /(N_F - N_W)$$

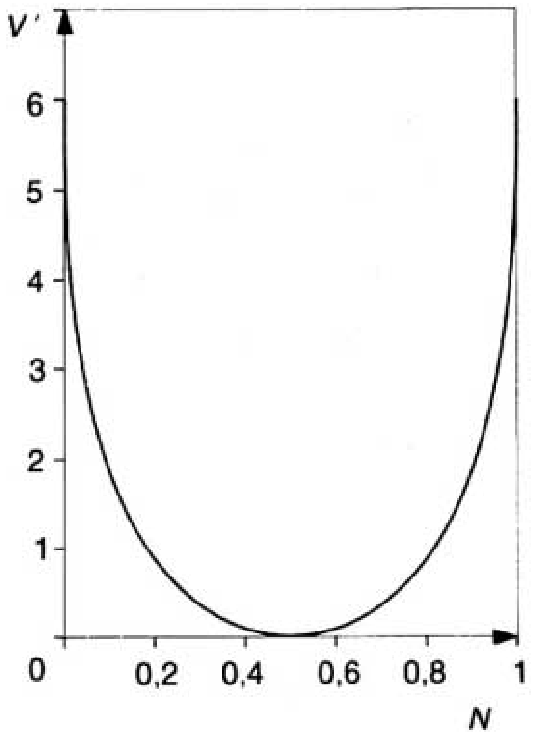


SWU, « separation working unit »

$$\mathbf{SWU = P.V(N_P) + W.V(N_W) - F.V(N_F)}$$

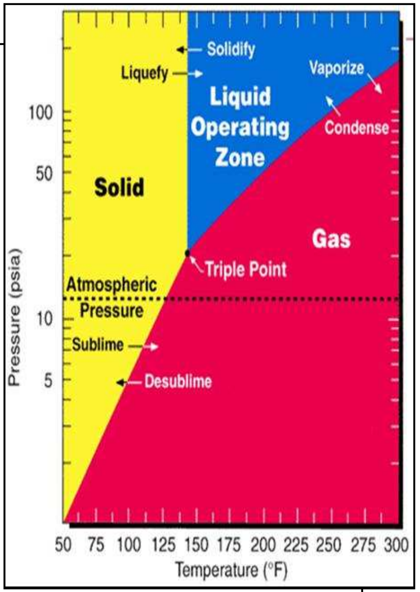
(V (N), « value function »)

$$\mathbf{V(N) = (2N-1)\ Ln\ [N / (1-N)]}$$



ISOTOPE SEPARATION PROCESSES

- **Nucleus mass ^{235}U vs.^{238}U:**
  - → *gaseous diffusion UF6 (GD)*
  - → *gaseous centrifugation UF6 (UCG)*
  - → *(balistic effects)*



- **Electron cloud properties ^{235}U vs.^{238}U:**
  - → *(chemical separation processes) (TC)*
  - → *(selective atomic ionization) (AVLIS)*
  - → *(selective molecular dissociation)*

GASEOUS DIFFUSION:

FROM ELEMENTARY ENRICHMENT TO ENRICHMENT CASCADES

PRINCIPLE: *UF_6 diffusion kinetics difference through very small pores (0.01µm)*

Thermal equilibrium: *same kinetic energy for all particules*

$E = 1/2\ M\ v^2$ >>> $v = (2E/M)^{1/2}$

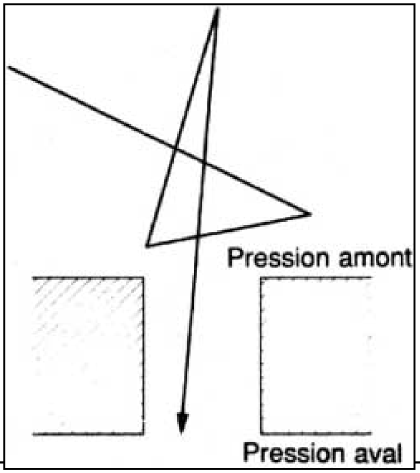
$M_{U235} < M_{U238}$ >>> $v_{U235} > v_{U238}$

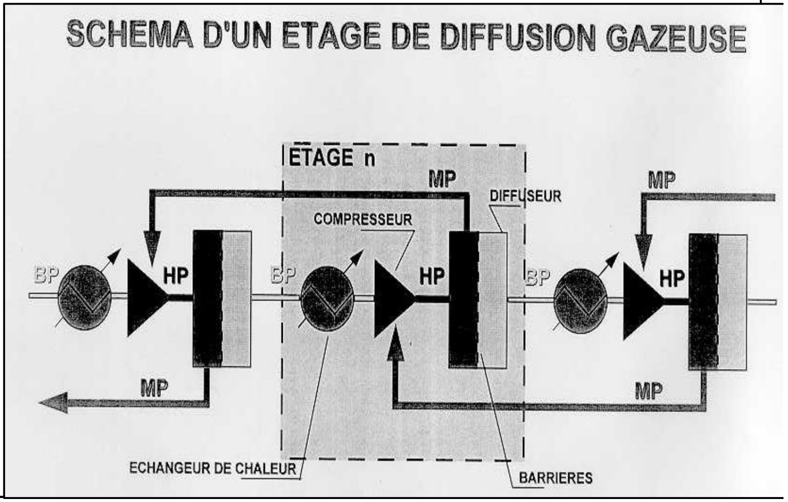
Enrichment factor:

$(M_{U238}/M_{U235})^{1/2} = 1.004$

[(theorical); in fact, lower...~1.002]

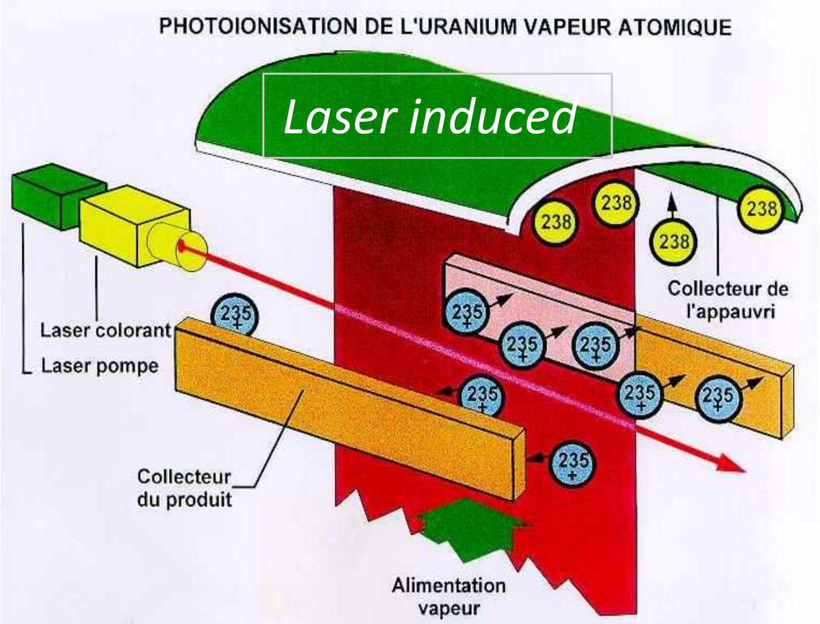
$(1.002)^{1000} = 7.3$



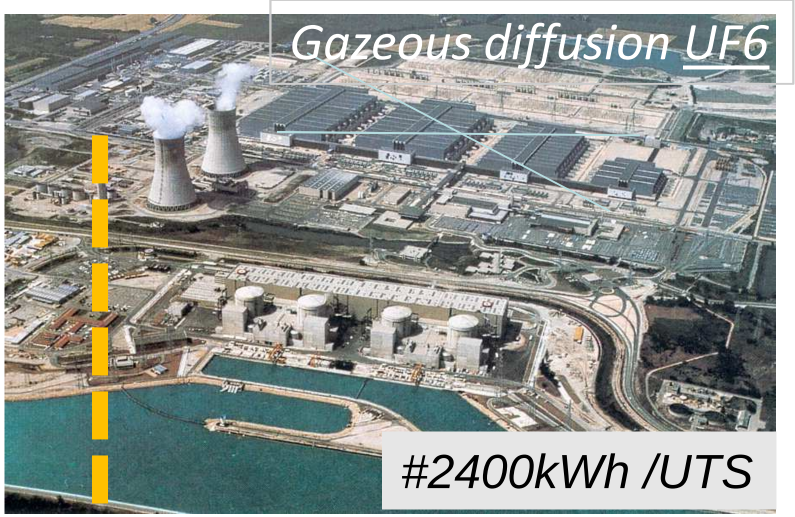


URANIUM ENRICHMENT PROCESSES

World fleet needs: ~ 50 MSWU

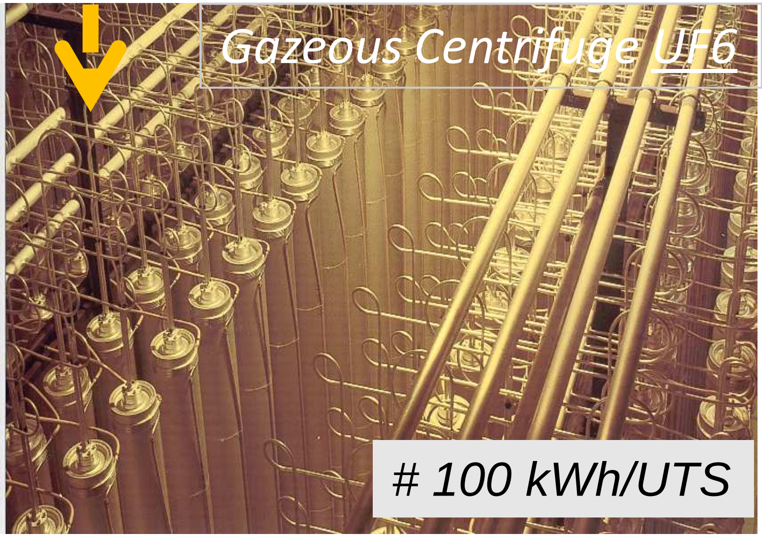


Laser induced



Gazeous diffusion UF6

#2400kWh /UTS



Gazeous Centrifuge UF6

 # 100 kWh/UTS

From UO3 to UF6:
the "CONVERSION" STEP

$$3\,UO_3 + H_2 \xrightarrow{450°C} 3\,UO_2 + 3\,H_2O$$

$$UO_2 + 4\,HF \xrightarrow{>150°\,C} UF_4 + 2\,H_2O$$

$$UF_4 + F_2 \longrightarrow UF_6$$