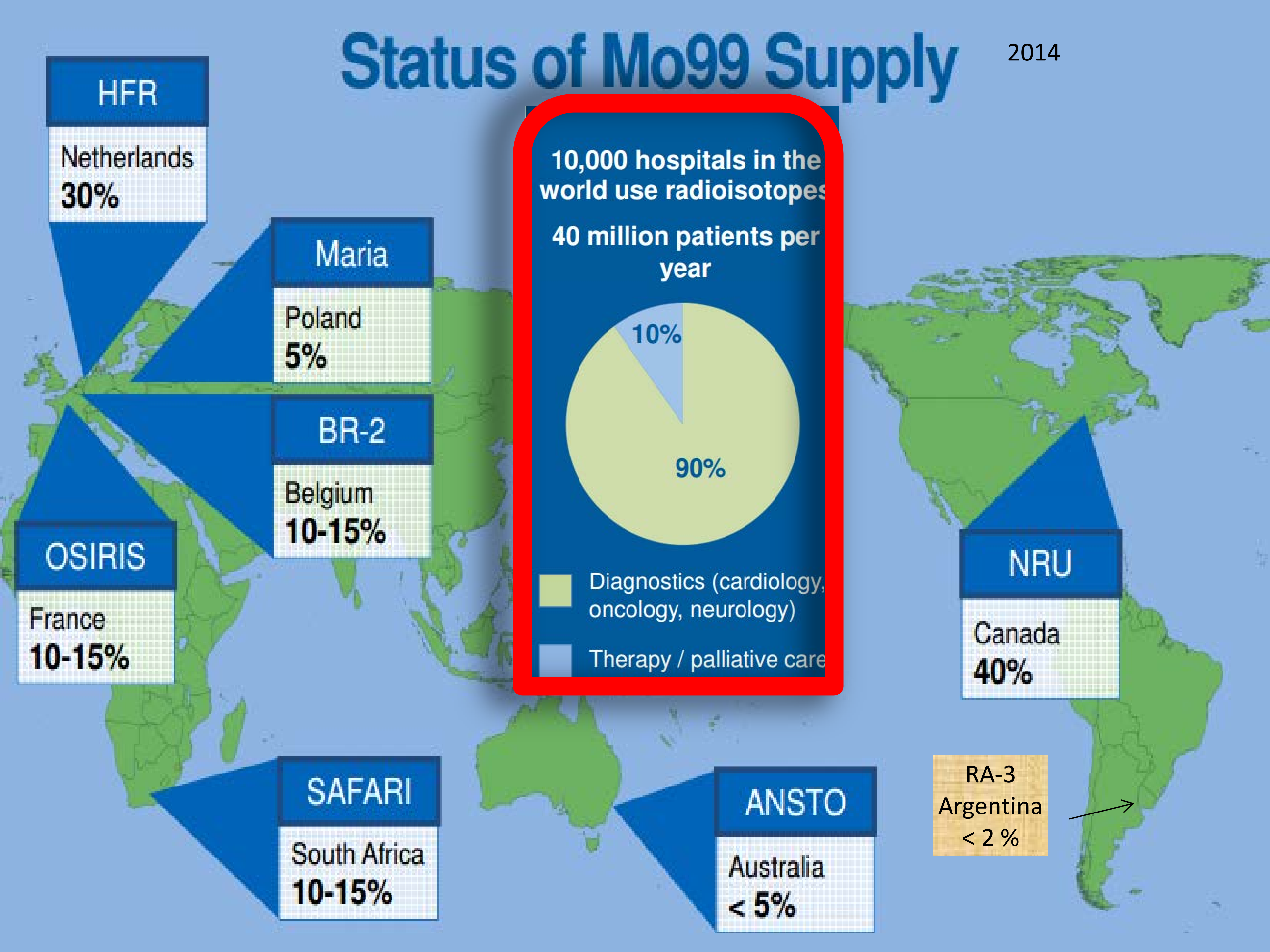
Irradiation Capsules with Suspended LEU UO_2 Particles for ^{99}Mo Production

E.E. Pasqualini, S. Navarro, G. Chetri, N. Gonzalez and J.C. Furnari

Nuclear Nanotechnology Laboratory
Constituyentes Atomic Center, CNEA.
Buenos Aires, Argentina

Mo-99 2016 Topical Meeting on Molybdenum-99 Technological Development
September 11-14, 2016. The Ritz-Carlton. St. Louis, Missouri


Status of Mo99 Supply
2014
HFR
Netherlands
30%
Maria
Poland
5%
BR-2
Belgium
10-15%
OSIRIS
France
10-15%
10,000 hospitals in the world use radioisotopes
40 million patients per year
10%
90%
Diagnostics (cardiology, oncology, neurology)
Therapy / palliative care
NRU
Canada
40%
SAFARI
South Africa
10-15%
ANSTO
Australia
< 5%
RA-3
Argentina
< 2 %

The homogeneous reactor concept applied to LEU ^{99}Mo production

Two types of homogeneous reactors

1) Dissolved uranium salts in acid solutions
2) Aqueous suspended uranium particles

Justification:

- ^{99}Mo from fission of LEU – *high specific activity*
- Fuel and target are the same – *high efficiency*
- Reutilization of ^{235}U without reprocessing – *high burn-up*
- Nuclear waste is reduced – *necessary tendency*
- Nearly 200 fold benefit – necessary *driving force to introduce technological changes*

Results:

- World provission of ^{99}Mo with a 2 MW reactor
- Interest in developing this idea

Drawbacks

Large ammount of solution to be processed

Alternatives:

Tendency to increase power density

- MIPS (1 kW/liter) U salts
- Robust MIPS (5 kW/liter) U salts

 # Need to move to suspended particles to reach power densities higher than 100 kW/liter, common in miniplate target irradiations.

 # Heat extraction compatibility

Aqueous suspension of LEU UO_2 particles in a closed capsule target as an intermediate step. POC

Overview **SPIM**

(Sistema de Producción de Isotopos Medicinales)

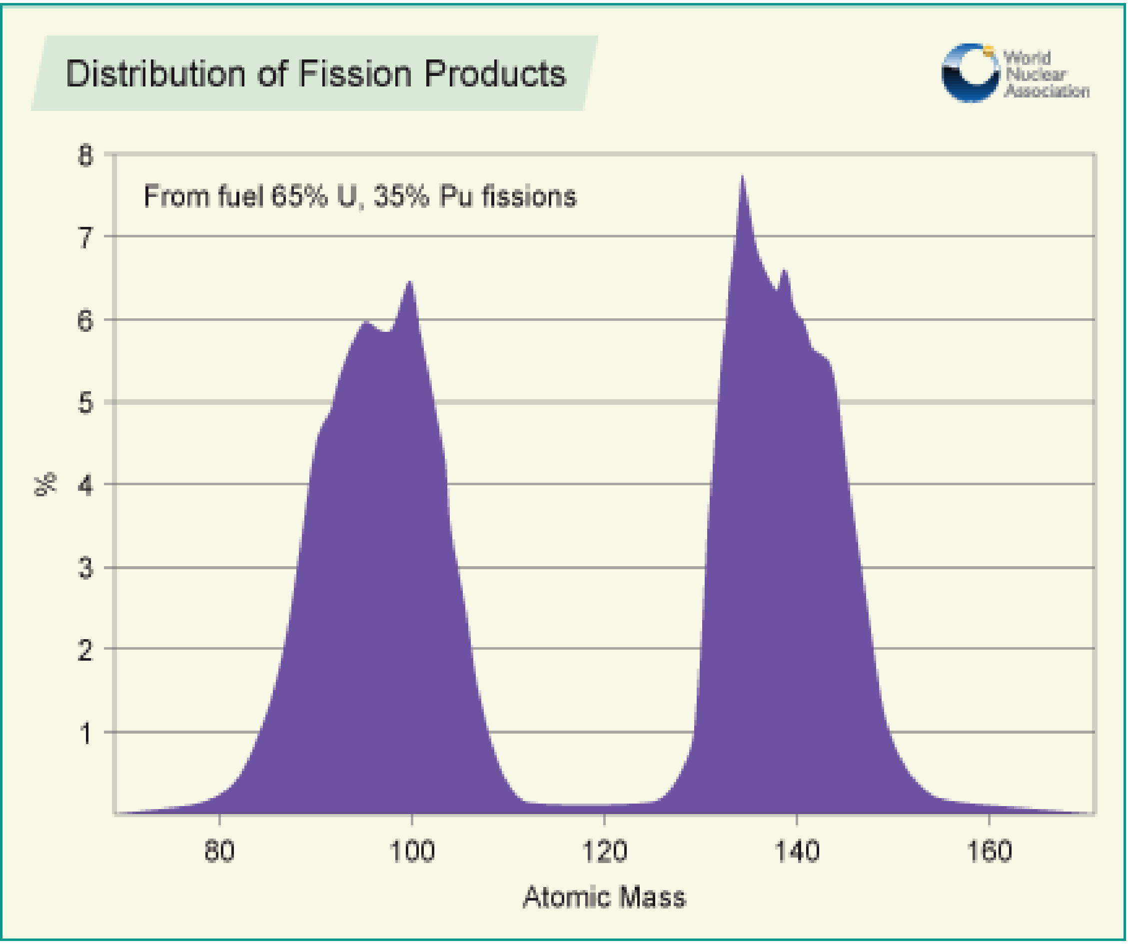
- Basic ideas
- Particles production
- Mo extraction experimental simulation
- Capsule characteristics
- Water radyolisis control
- Particles suspension
- Qualification

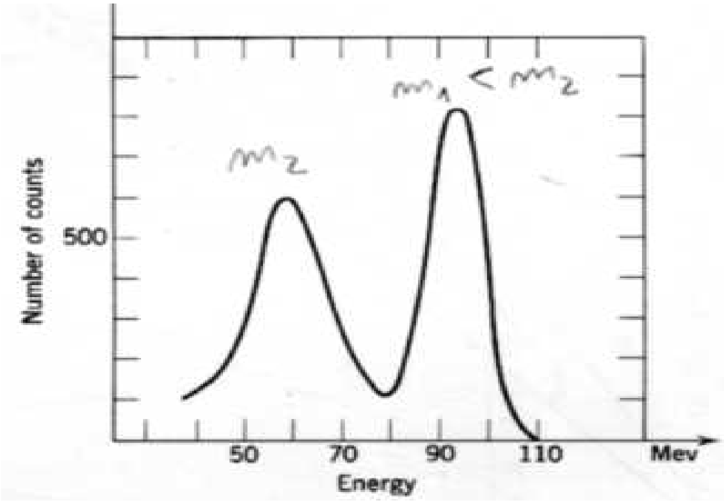
Isotopic distribution of ^{235}U fission products

^{235}U (n, x.n) A + B

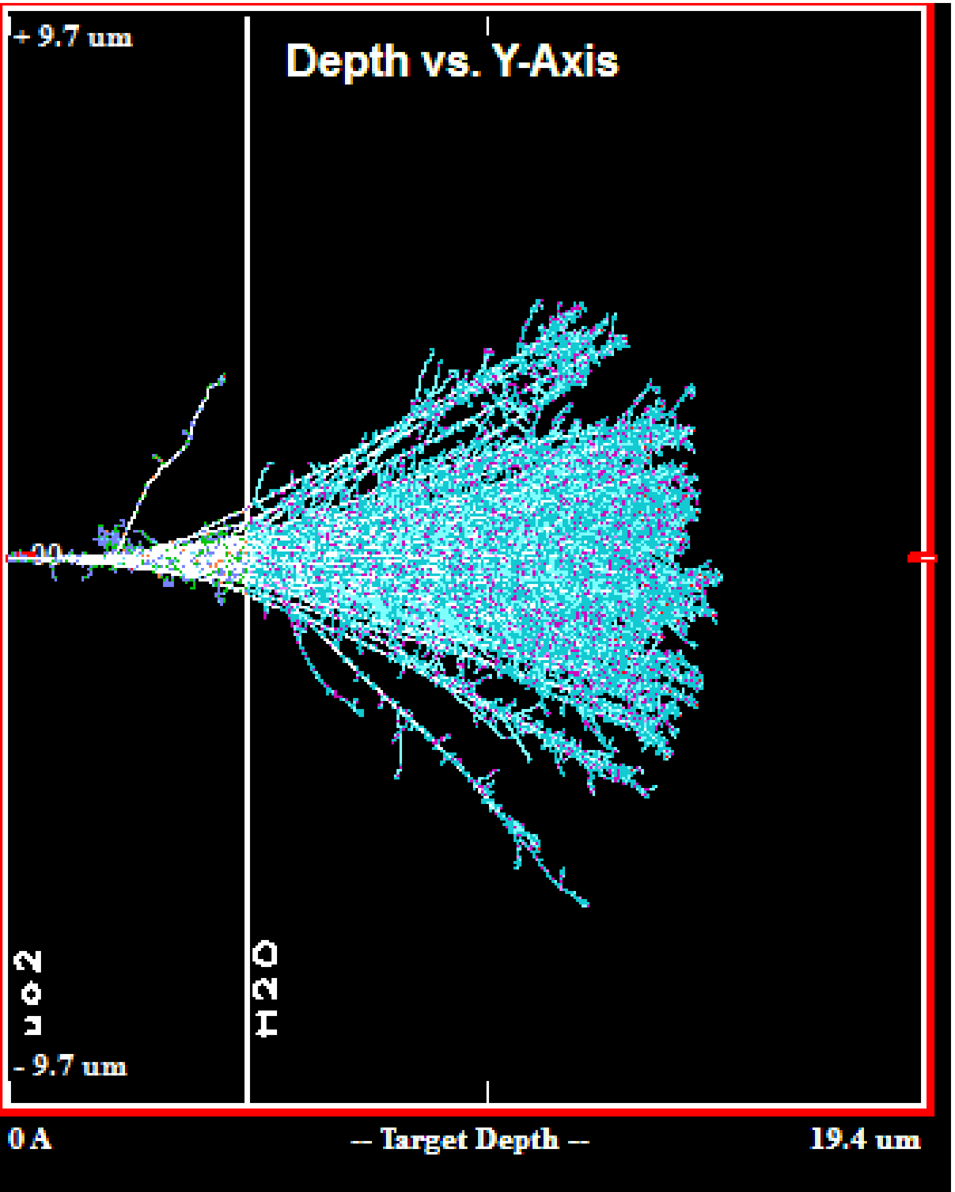
A between 83 and 106 amu
(E > 95 MeV)
Br, Kr, Rb, Sr,
Zr, Mo, Ru, Rh

B between 128 and 151 amu
(E > 60 MeV)
Sn, I, Sb, Xe, Cs,
Ba, Ce, Nd, Sm, Pm





Transport of Ions in Matter (TRIM)

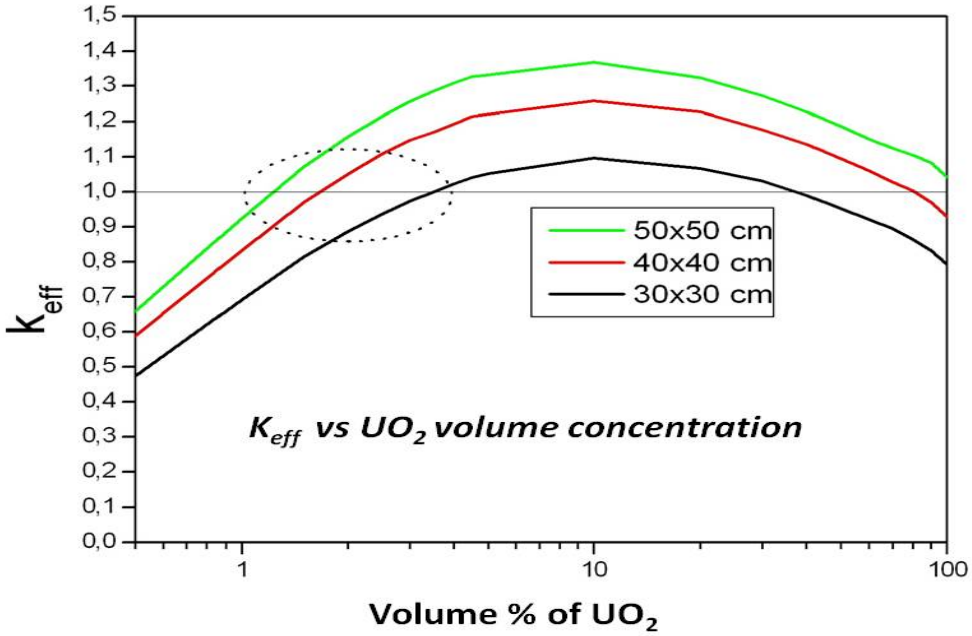


Trajectory of 1000 ^{99}Mo nuclides
with 90 Mev initial energy in
5 microns of UO_2
and final stopping in water

PENETRATION RANGE

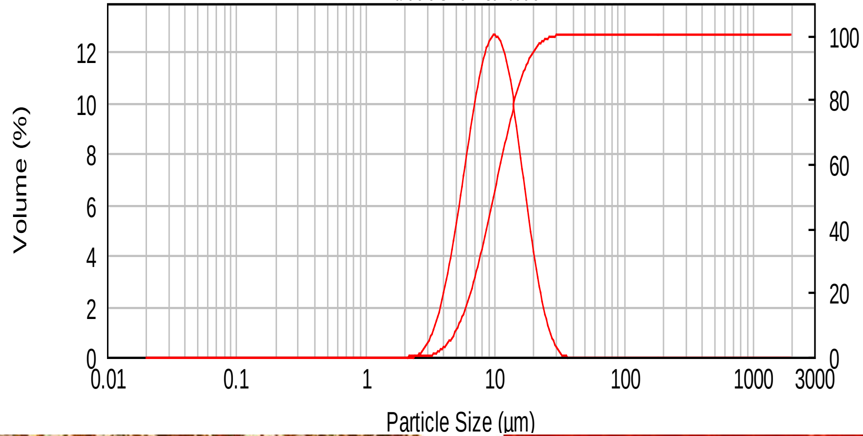
Isotope	Energy	UO_2	H_2O
	MeV	µ	µ
^{137}Cs	60	**5,73**	**18,0**
^{99}Mo	90	**7,15**	**21,2**

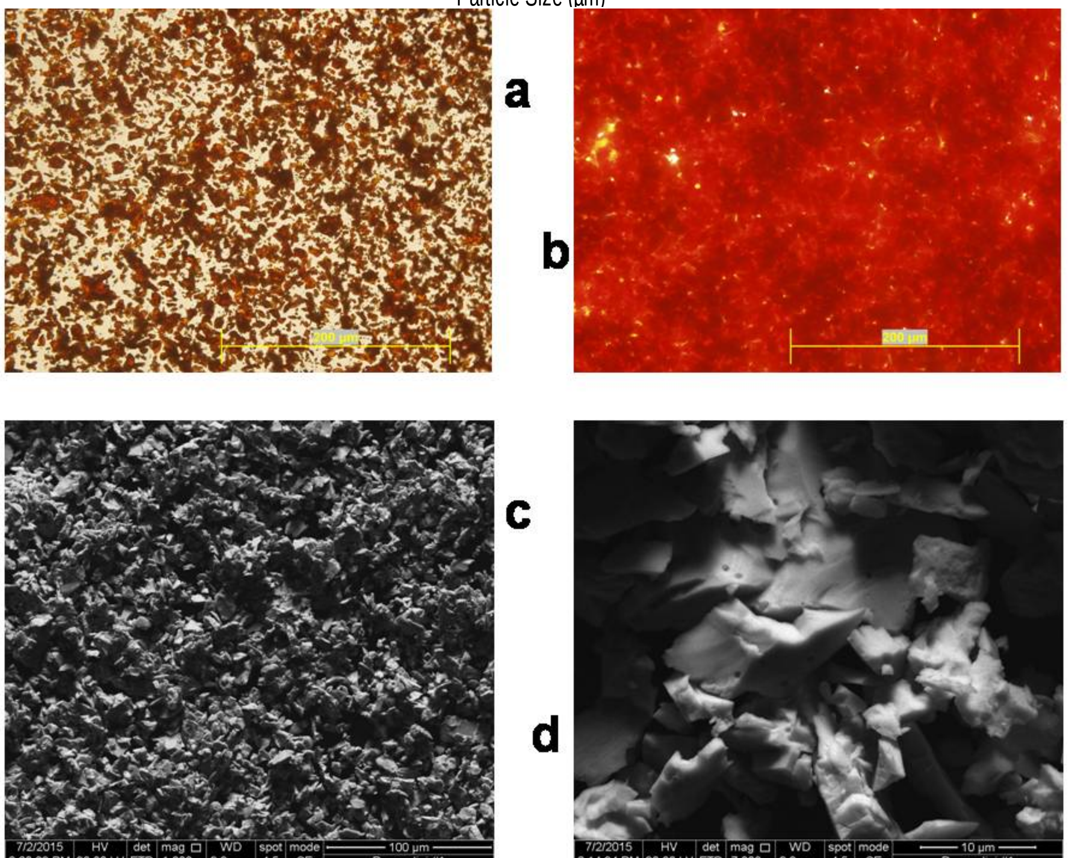
Suspended particles concentration for obtaining criticity in a nuclear reactor



Different cylindrical reactor sizes surrounded with 30 cm water reflector

 # Between 2 and 3 % v/v concentration is compatible with low fission products implantation in passing by particles



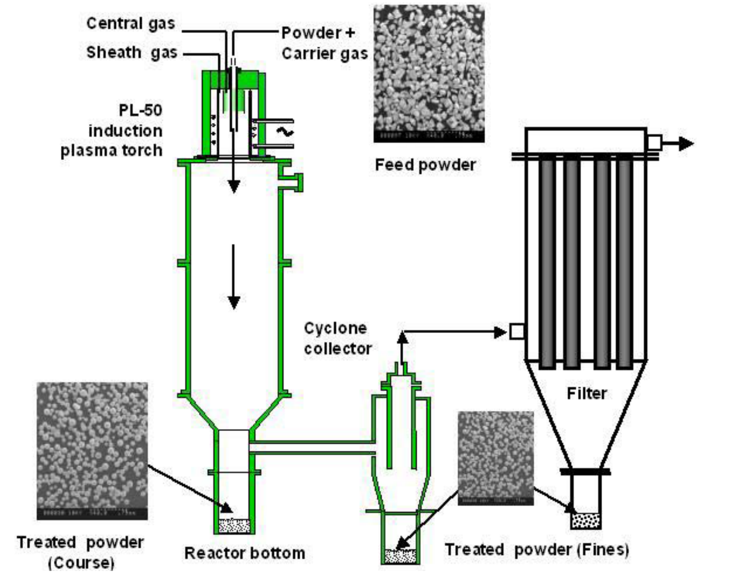


UO_2 *particles of10 µ obtained from pellets rectification and size separation*

a) y b): optical microscopy 800X . c) y d) SEM 1000 X y 7000 X ; S = 0,318 ± 0,003 m^2/gr

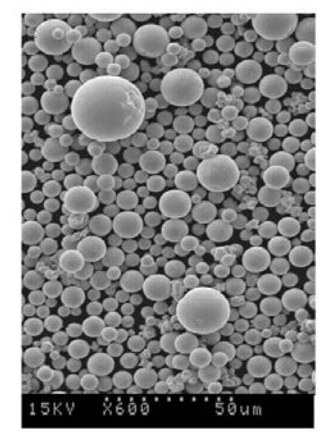
PARTICLES SYNTHESIS

6 ± 1 µ de diáameter



Spheroidization process scheme using inductive coupled plasma (ICP) using 2.5 MHz radiofrequency with a 15 kW power equipment (TEKNA, Montreal)
15 kW; 2,5 MHz

Cámara de esferoidización



Polvo esférico de tungsteno

ICP chamber and W spheroidisized powder

Particles spheroidization equipment

Inductive Coupled Plasma (ICP)* 2.5 MHz,15 kW. *TEKNA, Canada

Arriving at Constituyentes Atomic Center

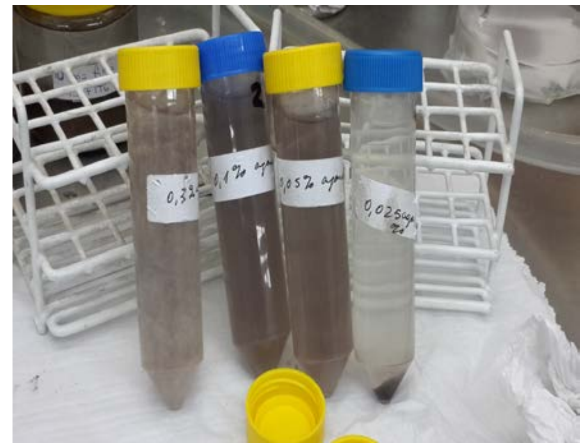
Positioned in the Lab

Cold adsorption of molybdate anion in UO_2 particles

Filtration or centrifugation desorption

Volume of suspensions = 50 cm^3

Filtering system for the desorption of MoO_4^{-2} from the UO_2 particles surface



Centrifugation tubes with suspended UO_2 particles in a tyxotropic solution of agarose

Separation processes of molybdate anion by filtration or centrifugation

Mo and U concentration of solutions in contact with UO_2 particles at different processing steps. Mo and U concentrations are measured with an error of 10%.

Procedure:		**Filtering**				**Centrifugating**	
Sample:		**[Mo] 5 ppm**		**[Mo] 0,5 ppm**		**[Mo] 1 ppm**	
Step	***Solution***	Mo (ppm)	U (ppm)	Mo (ppm)	U (ppm)	Mo (ppm)	Mo (ppm)
1	*Initial (Na_2MoO_4)*	5.8	< 0.1 ND	0.47	< 0.1 ND	1.2	< 0.1 ND
2	*First separation*	5.1 (88 %)	1.1	0.39 (84 %)	0.78	0.3 (25 %)	11
3	*Washing (NaOH)*	0.58 (10 %)	1.5	0.12 (26 %)	2.0	0.68 (57 %)	25
4	*Rinsing (H_2O)*	0.19 (3 %)	0.43	0.027 (6 %)	0.73	0.22 (18 %)	2,7

Planed irradiation in RA-1 reactor (10 kW)

Thermal neutron beam *(Φ = 4,5.10^7 n.cm^{-2}.s^{-1})*

11 gr of 10µ natural UO_2 particles (2% v/v)

In agarose solution (0,25 %)

Irradiation time	*Decay time*	*Activity*	*Contact dose*
Hours	*hours*	*µCi*	*µSv/hr*
8	16	5	100

ND temperature increment.

ND pressure increase (radyolisis or fission gases).

Gamma Spec measurements with HP Ge

of gas, liquid and solid after each separation step.

ND : not detected

PRECEDENT WORK
RERTR 1997
DISSOLUTION OF LOW-ENRICHED UO2/Al DISPERSION PLATES IN ALKALINE PEROXIDE SOLUTION (74-149 μ)

C. Conner, S. Aase, D. G. Wygmans, G. F. Vandegrift, D. Wu and S. Landsberger

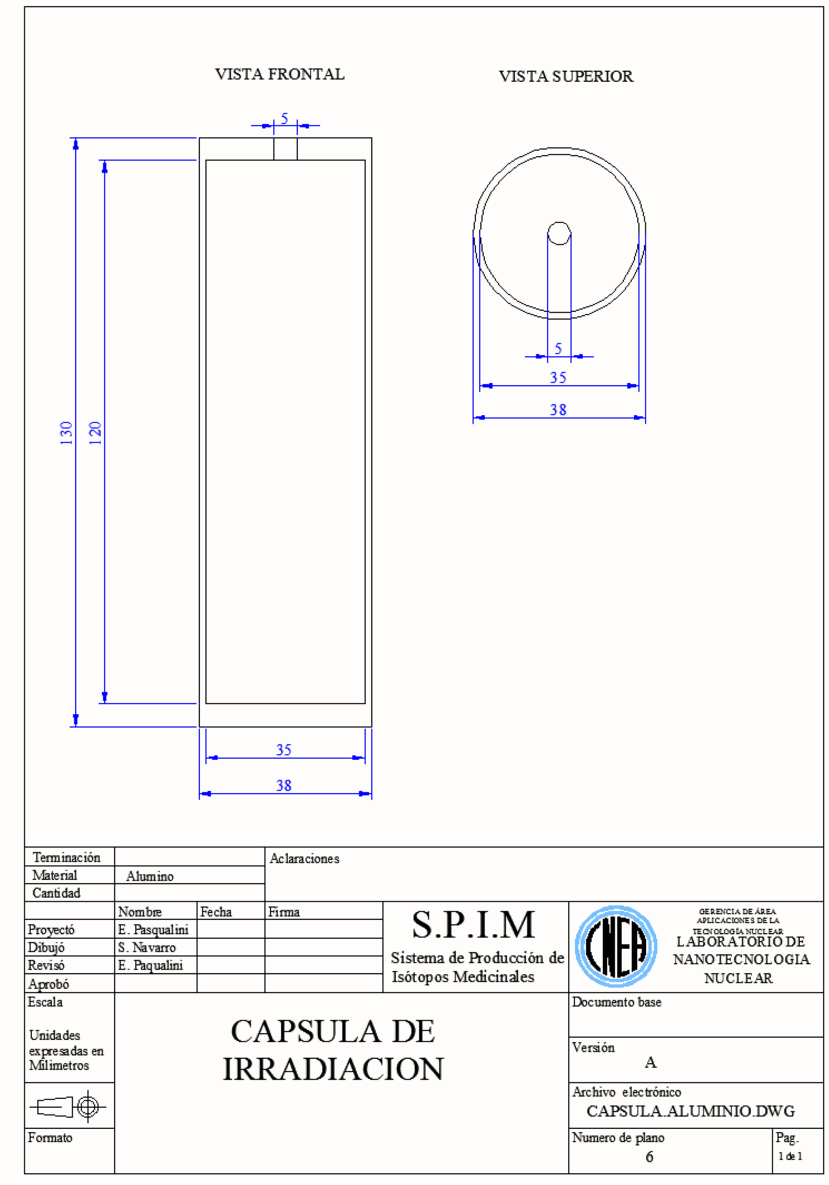
Table 1. Distribution of Uranium and Fission and Activation Products as a Percentage of the Initial Amount Present

Isotope	Aluminum Dissolution[a]		UO_2 Washings[b]		UO_2 Dissolution[c] (%)	H_2O_2 Destruction[d] (%)
	Liquid (%)	Solid (%)	Liquid (%)	Solid (%)		
Mo-99	5.9	0.5	0.2	1.4	92.0	83.8
U	0.1	2.0	0.1	6.8	89.1	1.9
Sr-91	0.4	6.9	0.2	1.7	90.8	N/A
Sr-92	6.1	1.2	0.2	1.3	91.2	N/A
Zr-95	5.6	1.5	1.1	2.1	89.7	3.6
Zr-97	5.6	0.9	0.2	1.4	91.9	N/A
Ru-103	6.8	1.5	0.3	1.3	90.1	79.4
Te-132	6.4	1.6	0.4	1.6	90.0	1.3
I-131	6.9	1.3	0.2	1.3	90.3	68.7
I-133	6.3	1.4	0.2	1.3	90.9	66.2
I-135	6.1	1.1	0.3	1.8	90.8	N/A
Ba-140	0.6	5.3	0.1	1.8	92.1	<4.1
La-140	0.6	7.4	0.1	1.7	90.2	<5.7
Ce-141	0.4	6.5	0.1	2.0	91.0	<5.6
Ce-143	0.5	5.7	0.0	1.9	91.9	0.8
Np-239	0.0	0.2	0.0	1.6	98.2	21.2

High power density irradiation capsule

Stainless steel.
Less than 0.5 mm thick.

Can be adapted to different reactors.
Specially to RA-3 where ^{99}Mo is produced In Argentina.
Also can be used In the RA-10 in construction,
Having similitudes With the Australian OPAL Rector.

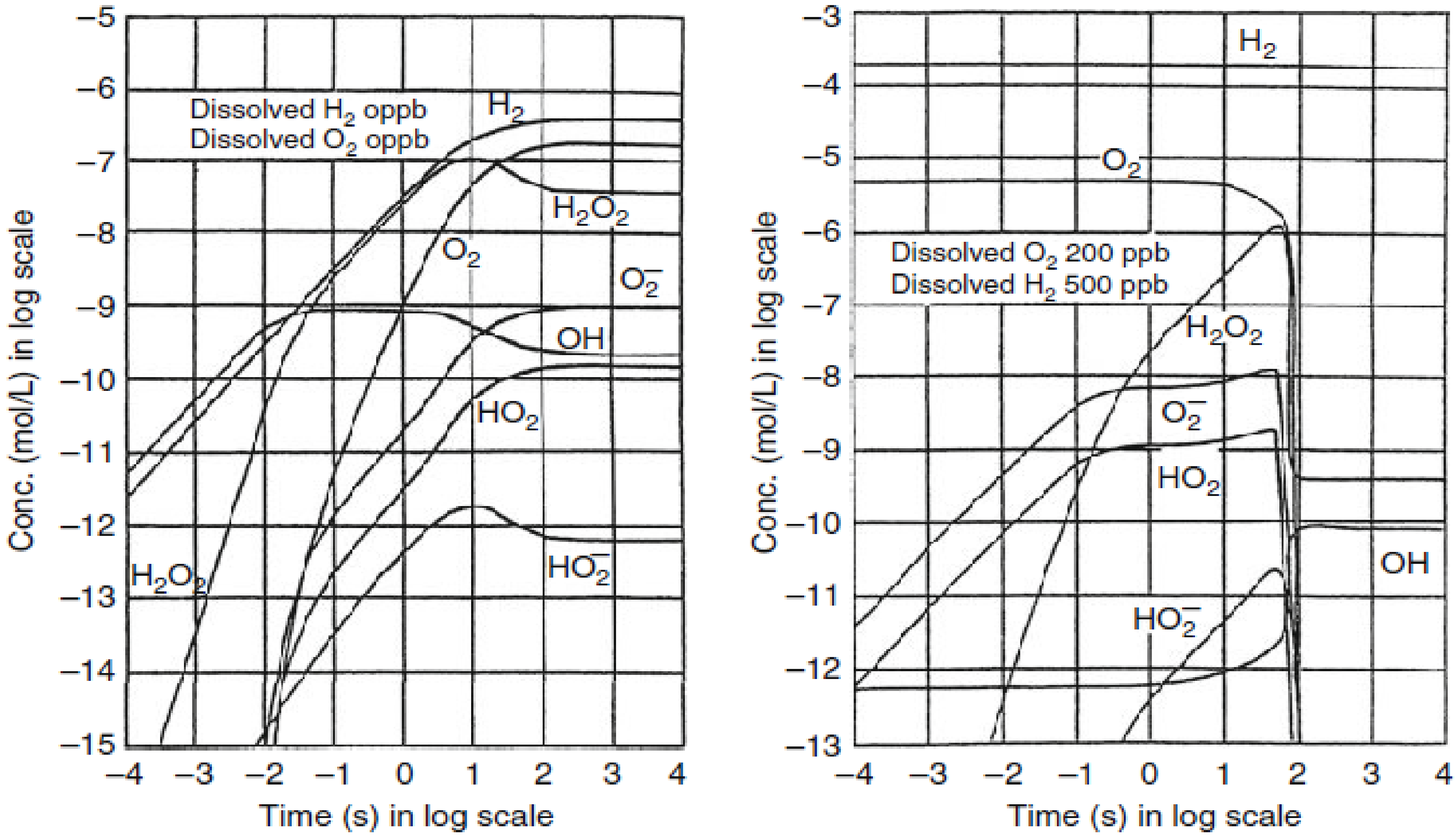


<u>Capsule characteristics</u>

- Filter inside the capsule
- 10 % empty suspension volume
- Hydrogen presurized up to some atmospheres
- Thermal neutrons flux <u>8 x 10^{13} n/cm^2s</u> (RA-3)
- Total ^{235}U = 5 gr; 28,5 gr of UO_2 (LEU)
- Total volume of particles = 2,6 cm^3; aprox. <u>2,5 % v/v</u>
- Total power = 10,6 kW (3,3 x 10^{14} fisiones/seg); <u>92 kW/lt</u>; 80 W/cm^2
- Fission gases pressure increment after 7 days irradiation = 85 mbar
- EOP = <u>500 Ci</u> de ^{99}Mo (irradiación 7 días)
- Consumed ^{235}U per cycle = 1,5 % (<u>reciclable</u> > 50 % burn-up)
- Entrances to facilitate automation processes of liquid extraction, washing and rinsing.

Water radiolysis control

- Lowering equilibrium recombination pressure with hydrogen incorporation
- Mean energy deposited directly in the water = 70 % of fission energy with a LET of 3800 keV/µ.
- Total power is 7,5 kW y a water volume of cm^3.
- Values to be used for the calculation of initial hydrogen incorporated pressure

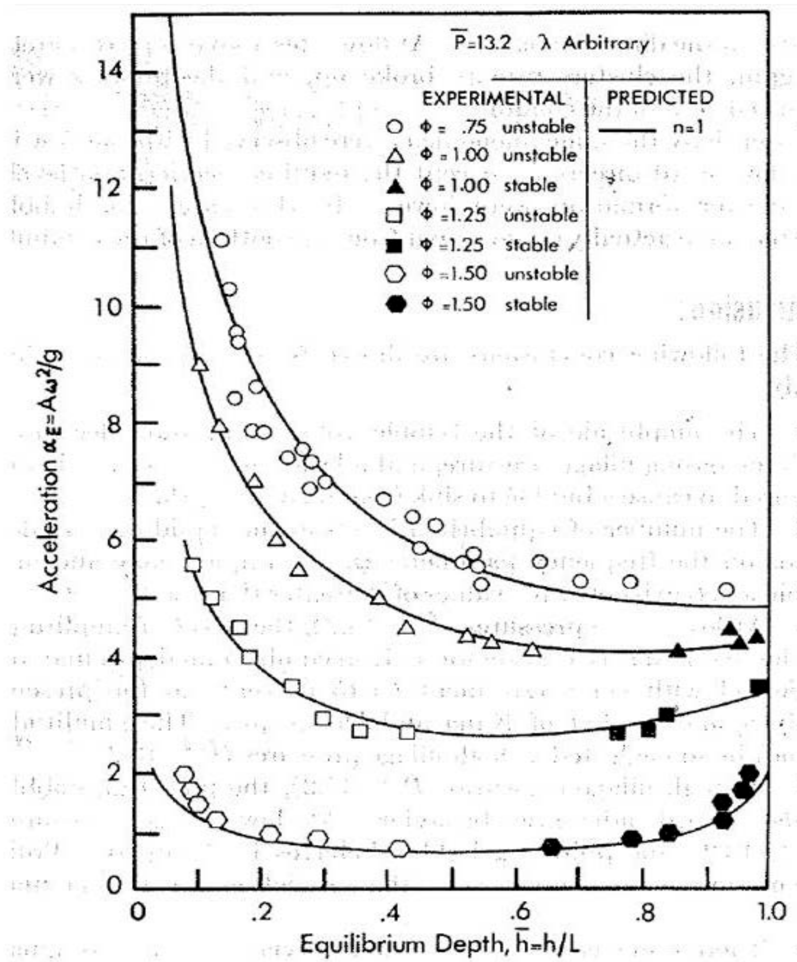


Radiolysis computational simulation showing at right the pressure equilibrium descent with hydrogen incorporation

Particles suspension

100 Hz frequency, amplitude 5 mm p/p
dos steps 1) Surface bubles formation 2) Bubbles dipping

Transparent capsule in a 11 kN shaker



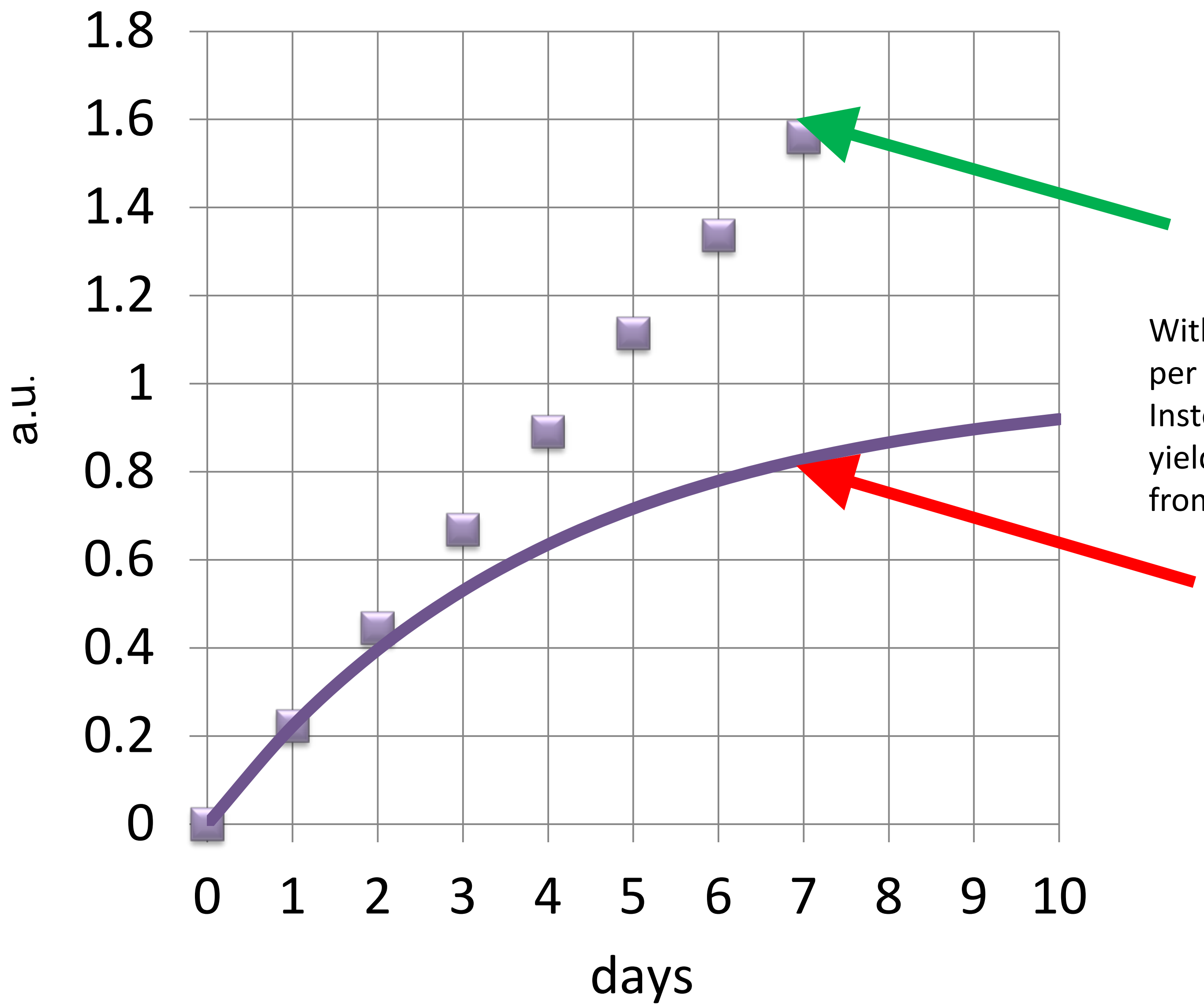
Bubbles movement in a liquid column within an oscilatory field of low frequency

VIBRATOR

ONLY WATER

UO_2 WATER SUSPENDED

^{99}Mo: Continuous accumulation vs. daily extraction.



With one extraction per day of Mo-99, Instead of weekly, yields can be doubled from 0.8 to 1.6.

PERSPECTIVES

- Innovative project
- Needs of proof of concept
- Laboratory licencing for particles production and analysis
- Adquired equipment for melted spherical UO_2 particles preparation
- First irradations in a few months
- Out of pile capsule qualification
- Easy automated possibilities
- Expected high production yields
- Studies on nuclear reactor parameters
- The capsule target can be used with any neutron producing source.

Thanks