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Molten Salt Reactors: Innovative Designs and Calculations of MSR Neutronics

Joint ICTP-IAEA Workshop on Physics and Technology of Innovative High Temperature Nuclear Energy Systems

14-18 October 2019

ICTP, Miramare - Trieste, Italy

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The Stage: McMaster University

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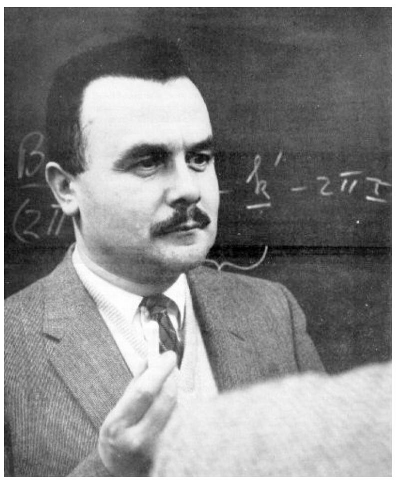
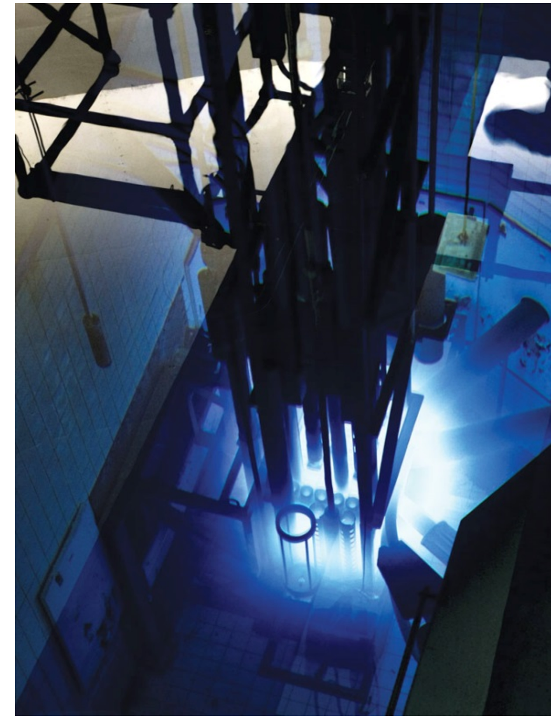
- **McMaster Nuclear Reactor** Critical **April 1959** (First RR at a Commonwealth University) (CERN:1952)
- **Bertram Brockhouse** shared the 1994 Nobel Prize in Physics with American Clifford Shull for developing neutron scattering techniques for studying condensed matter.



Today: McMaster Research Funding about $400M – one of Canada’s most research intensive Universities

MNR:
- Intense positron beam
- Small-angle neutron scattering
- Neutron activation analysis
- Neutron radiography



MNR: Commercial production of radio-isotopes for medical purposes (I-125, Lu-177, Re-186, …)
Accelerators (F-18), Hot cells, Sources.
https://nuclear.mcmaster.ca/

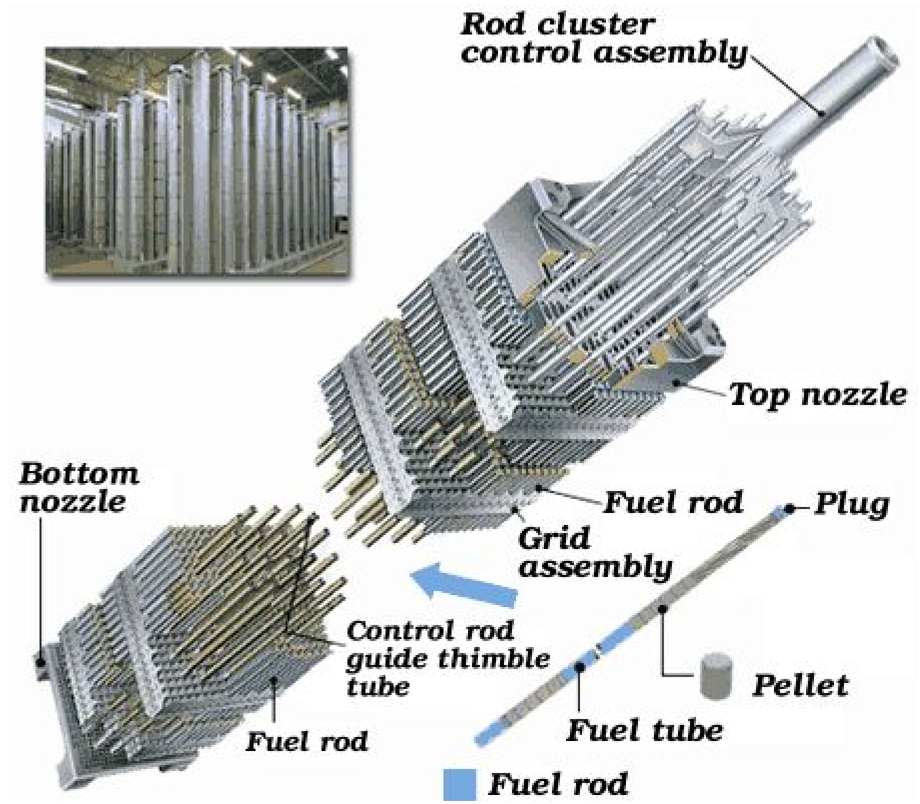
2018 Nobel Prize Donna Strickland was student at McMaster

Outline

- The idea behind molten salt reactors
- History of molten salt reactors
- Introduction to (relevant) neutronics
- Neutronics of molten salt reactors
- Current designs of molten salt reactors

Burnup distribution

- Fluxshape (power profile):
  - Axial ?
  - Radial ?
- Need to shape the flux
  - Graded enrichment
  - Control devices
  - (burnable absorbers)
  - Fuel shuffling between reloads:
    - Radially (PWR, BWR)
    - Axially (PHWR)
- Always uneven burn-up
  - But jobs for engineers!



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Liquid fuel

- Imagine you could use liquid fuel, flowing through the core:
  - Flux shape (power profile) would still be the same:
    - Axially: $\sim\sin\left(\frac{\pi}{H}z\right)$ H is height of cylinder
    - Radially: $\sim J_0\left(\frac{2.405\,r}{R}\right)$ R is radius of cylinder
  - Burnup would be completely uniform!
    (provided there is perfect mixing)
- Other immediate advantages:
  - No core-meltdown! (semantics, it's molten already…)
  - No fuel failure
  - Fission gases can be vented off.
  - Fuel is the coolant, no coolant needed (in primary circuit).

Choice of Liquid (Fluid) Fuel

- Salt

- Wikipedia: a salt is an ionic compound that can be formed by the neutralization reaction of an acid and a base.
  Salts are composed of related numbers of cations (positively charged ions) and anions (negative ions) so that the product is electrically neutral (without a net charge).
- Salts characteristically have **high melting points**.
- Long list of requirements for fuel:

Liquid Fuel Requirements

- Low capture x-sec for neutrons (*)
- Stable against radiation (*)
- Needs to be able to dissolve enough fissile/fertile material to achieve criticality (*)
- Thermally stable (Eutectic)
- Low vapor pressure
- Good heat transfer
- Non-aggressive to structural components

(*) means relevant to neutronics

Choice of Liquid Fuel

- Only low-Z materials remain for neutronic reasons: Be, Bi, B-11, C, D, F, Li-7, N-15, O. (→ NNDC)
- Chemistry places additional requirements rejecting Bi, B-11, C, D, N-15, O;
- We are left with: F, Li-7, Be, commonly referred to as **Flibe**.
- Beryllium also acts as a neutron-doubler:

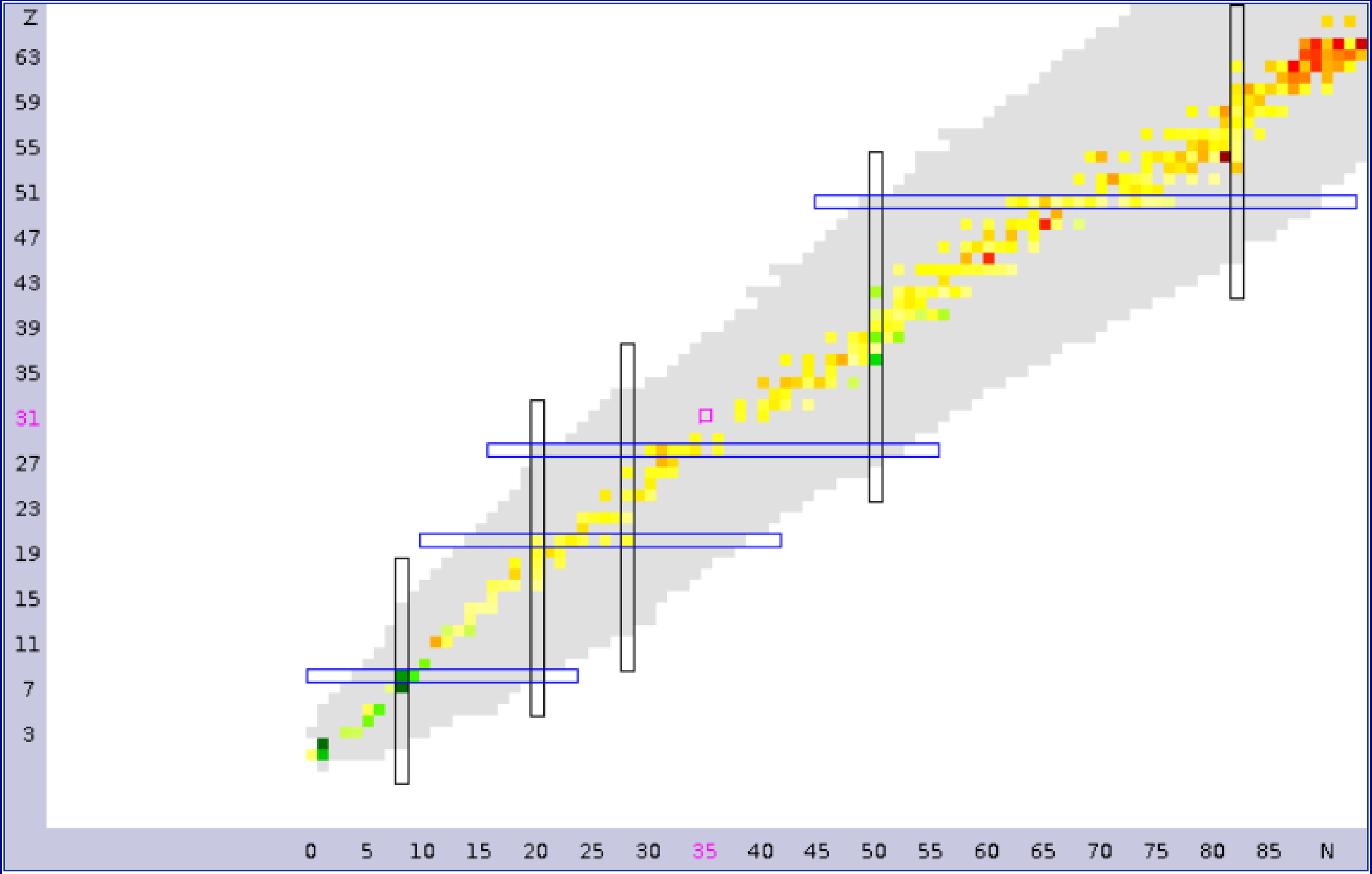
$$ {}^{9}_{4}\mathrm{Be} + n \rightarrow 2\,{}^{2}_{2}\mathrm{He} + 2n $$

- Also high elastic cross section → good moderator.
- But beryllium is poisonous.
- Other elements such as Zr, Na, K are sometimes added for different purposes.

Chart of Nuclides

Click on a nucleus for information

Color code	Half-life	Decay Mode	$Q_{\beta-}$	Q_{EC}	$Q_{\beta+}$	S_n	S_p	Q_α	S_{2n}	S_{2p}	$Q_{2\beta-}$	Q_{2EC}	Q_{ECp}
$Q_{\beta\text{-}n}$	BE/A	(BE-LDM Fit)/A	$E_{1st\ ex.\ st.}$	E_{2+}	E_{3-}	E_{4+}	E_{4+}/E_{2+}	β_2	$B(E2)_{42}/B(E2)_{20}$	$\sigma(n,\gamma)$	$\sigma(n,F)$	235U FY	239Pu FY



Tooltips
On
Off

Zoom
1
2
3
4
5
6
7

Uncertainty
NDS
Standard

Screen Size
Narrow
Wide

Nucleus

go

barns

1.01E+7	1.56E+1
2.66E+6	4.09
6.98E+5	1.07
1.83E+5	2.81E-1
4.80E+4	7.38E-2
1.26E+4	1.93E-2
3.30E+3	5.07E-3
8.66E+2	1.33E-3
2.27E+2	3.49E-4
5.95E+1	9.15E-5
1.56E+1	2.40E-5

unknown

Ground and isomeric state information for $^{66}_{31}Ga$

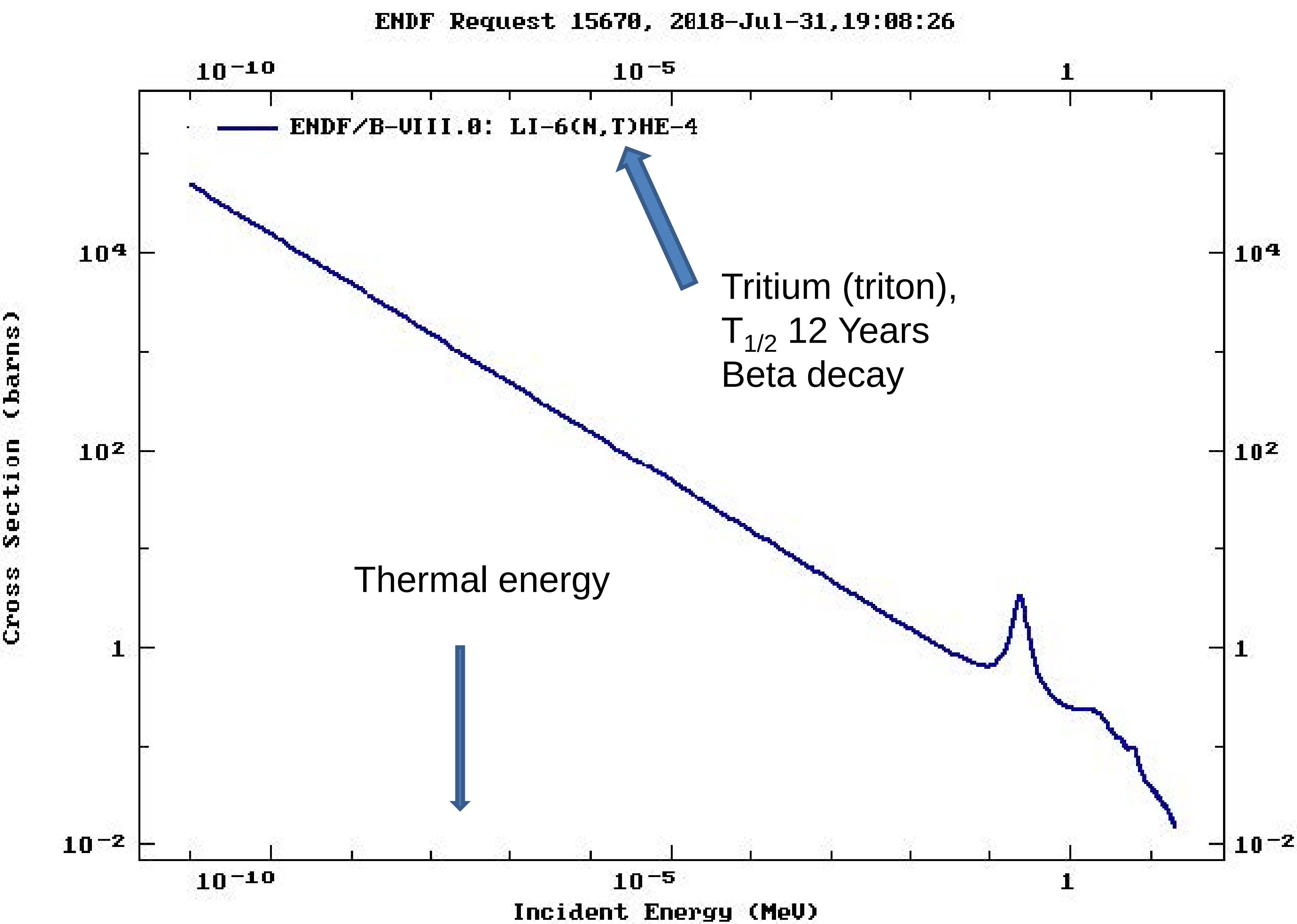
E(level) (MeV)	Jπ	Δ(MeV)	$T_{1/2}$	Decay Modes	σ(n,γ) (b)
0.0	0+	-63.72366015625	9.49 h *3*	ε : 100.00 %	

Search options:

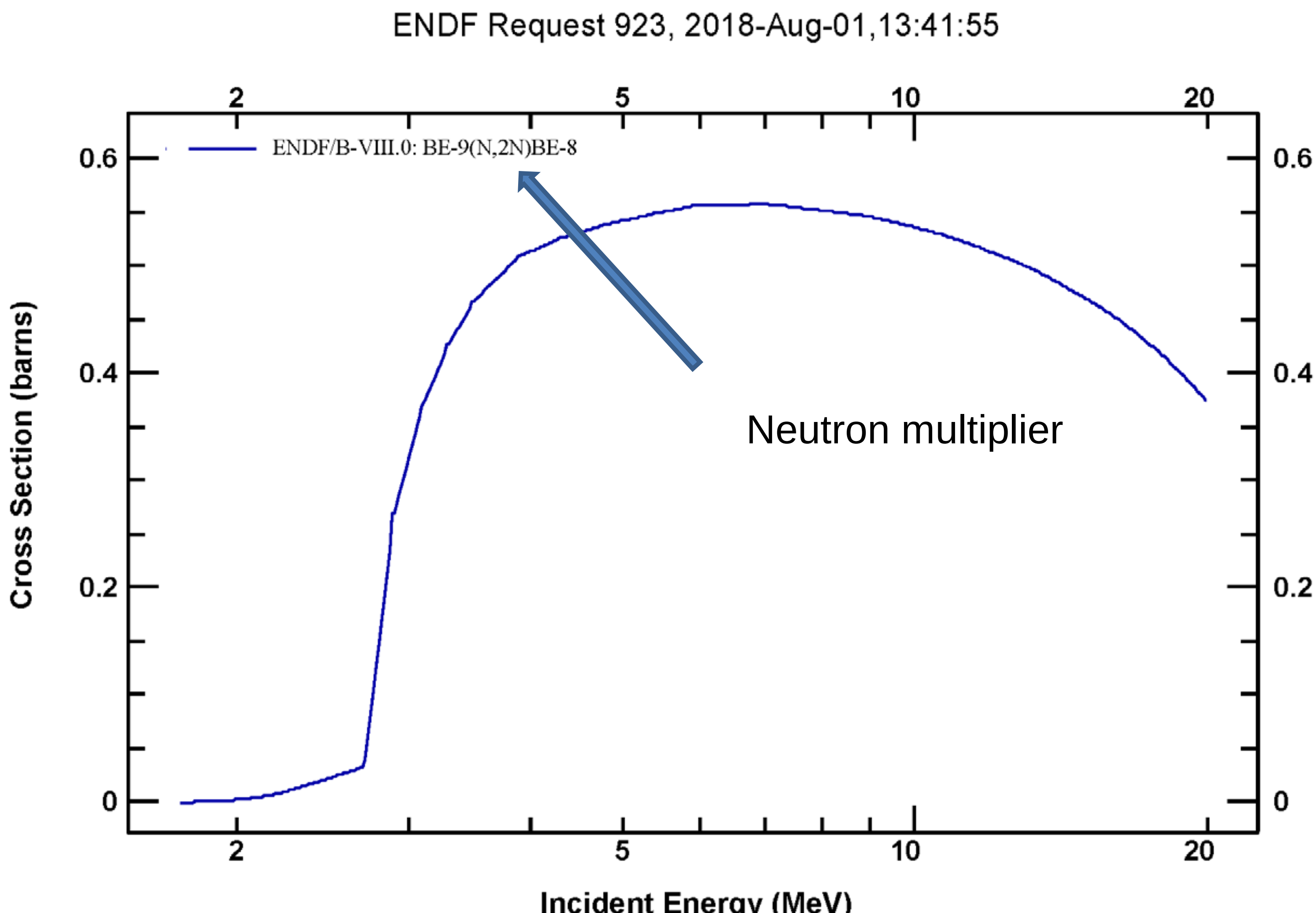
Levels and Gammas

Nuclear Wallet Cards

Li-6 Cross Section



Be-9 Cross Section



Fuel Salt

- Nuclear fuel is U, Pu, Th.
  (fissile, fissionable and fertile)
- Included in the salt as fluorides:
  - UF_4, not to be confused with UF_6, used in uranium enrichment process.
    - Uranium is enriched (typically 20%, LEU)
  - ThF_4, breeding material,
    - either in fuel or blanket.
  - PuF_3
- Typical salt would be (MSRE):
  - 65% ^7LiF – 29.1% BeF_2 – 5% ZrF_4 – 0.9% UF_4
  - With 35% enriched uranium

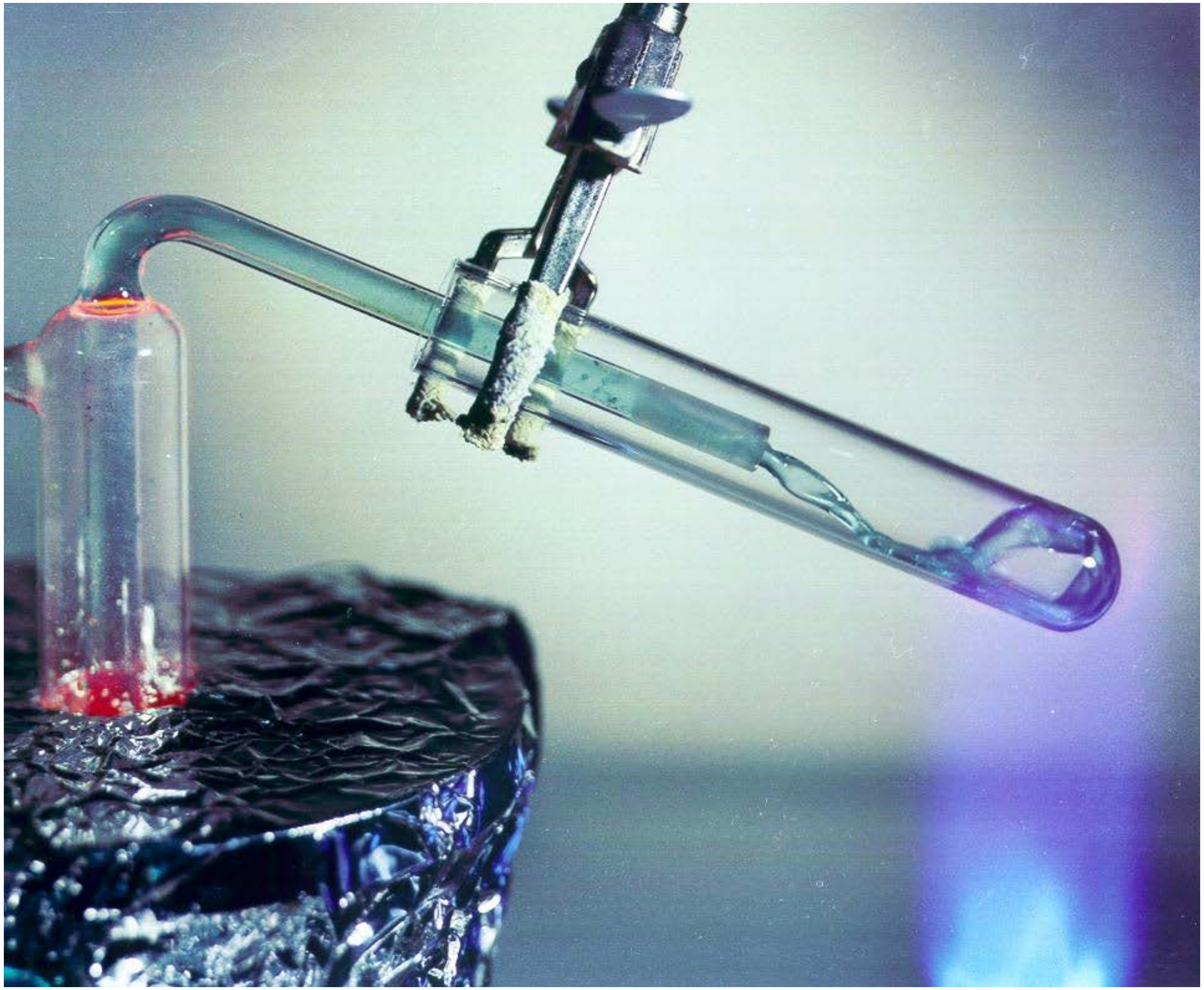
Fuel Salt Properties

MSRE Fuel

Property	H_2O	Na	Li	$^7LiF-BeF_2-ZrF_4-UF_4$ 65-29.1-5.0-0.9
Melting point (°C)	0	98	181	434
Boiling point (°C)	100	880	1342	1435
Density (kg/m^3) (*)	712	830	483	2300
Thermal conductivity (W/K/m) (*)	0.54	67	53	1.43
Specific heat capacity (J/g/K) (*)	5.7	1.26	4.23	2.0
Viscosity (10^{-6} Pa s) (*)	89	250	360	8050

(*) typical reactor conditions

Flibe



Strong Point of MSR

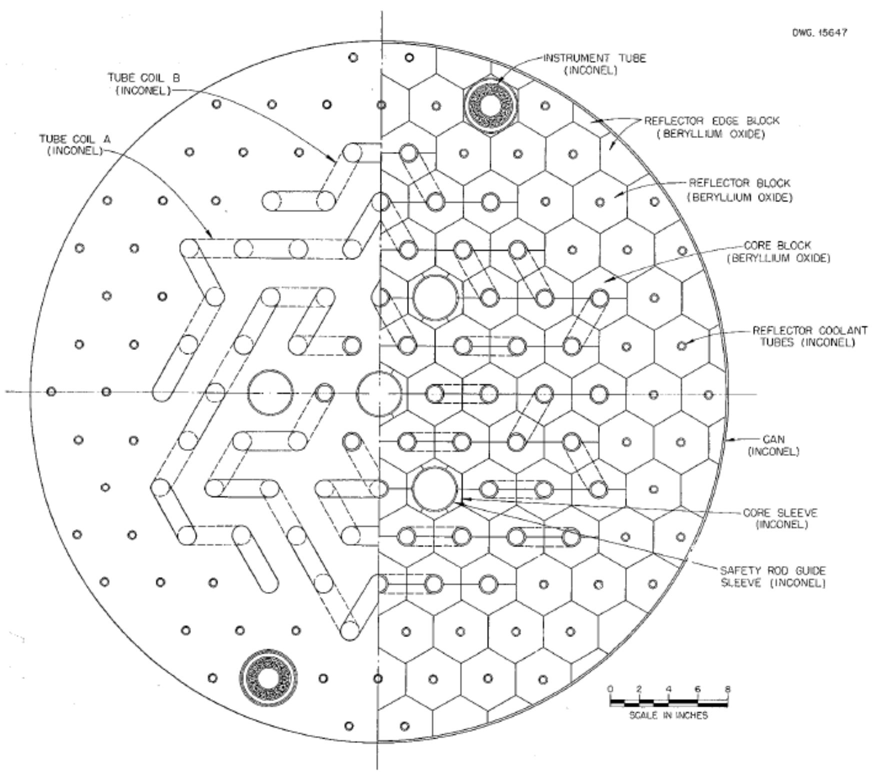
- Inherent safety:
  - No meltdown;
  - Negative power coefficient (*);
  - Dump tank with freeze plug;
- Fission products can be removed easily.
- Fission products form stable fluorides.
- Operation is at low pressure.
- Xe can be skimmed off. (*)
- Fuel can be added at will. (*)
- No water or sodium present, less risk of steam explosions or hydrogen production.

History

- MSRs were pioneered at Oak Ridge National Labs, Tennessee in the 1940`s
- First experiments were Aircraft Reactor Experiments:

Aircraft Reactor Experiment

- Operated for 9 days in 1954 (ORNL)
  - Salt: 53% NaF – 41% ZrF_4 – 6% UF_4 (HEU 93.4%)
  - Moderator: BeO, Temperature: 860 °C
  - Power: 2.5 MWth



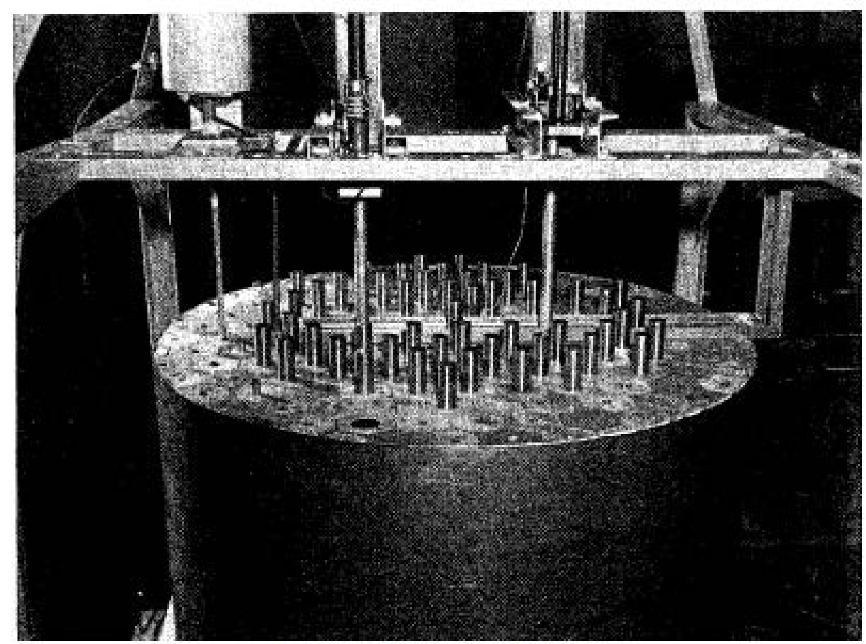


FIGURE 1: Critical Assembly of ARE

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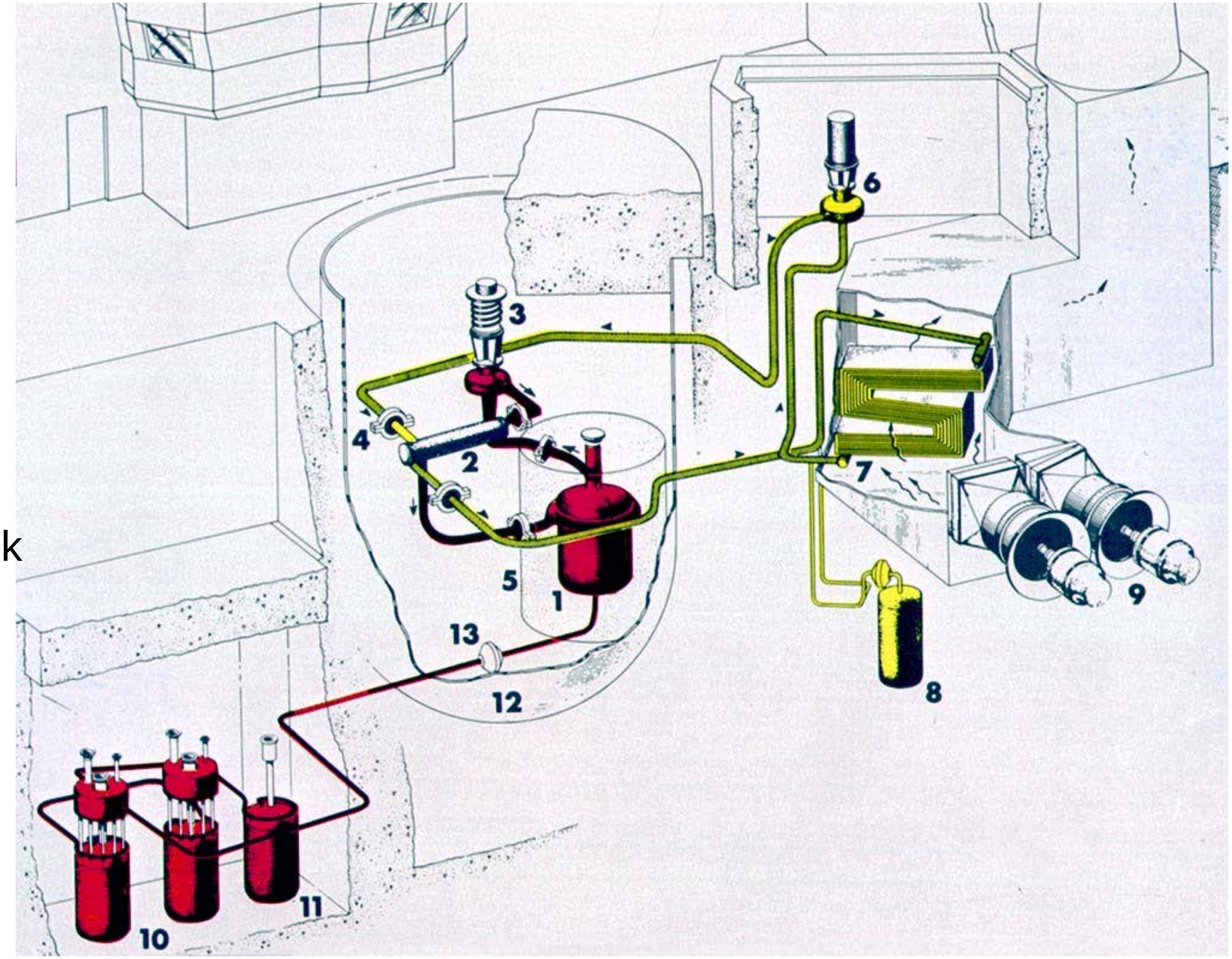
Molten Salt Reactor Experiment

- Operated from 1965 – 1969 (ORNL)
  - Salt: ^{7}LiF - BeF_2 - ZrF_4 - UF_4 (65- 29.1- 5 - 0.9)
  - 33% Enrichment. (^{233}U and ^{239}Pu also used)
  - Secondary circuit: LiF-BeF_2 (66–34 mole %)
  - Power 8 MWth, Temperature: 650 °C
  - Operated 9005 fph with U-235
  - Operated 4157 fph with U-233

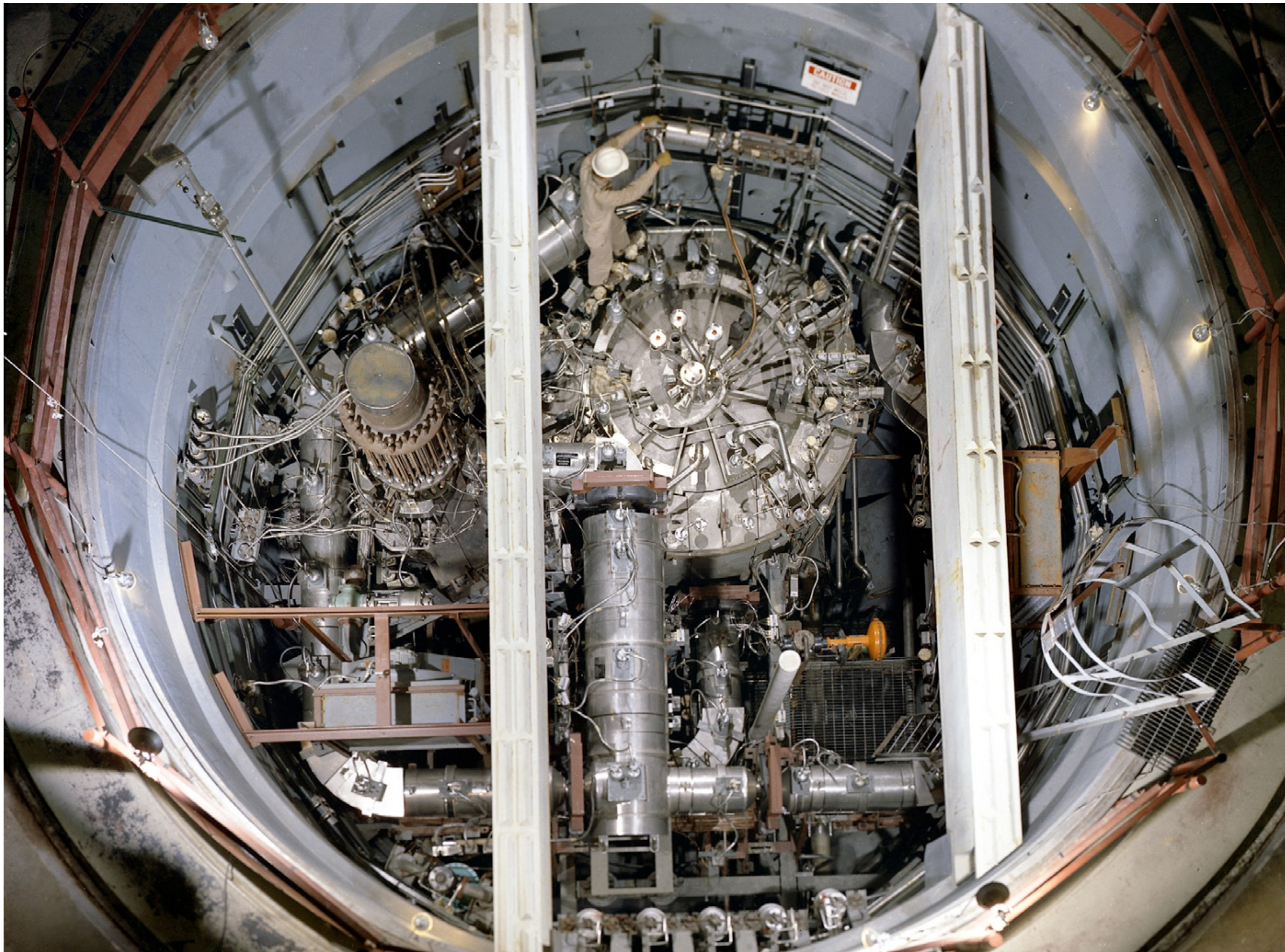
- It was a successful proof of concept

MSRE

1 Reactor vessel
2 Heat exchanger
3 Fuel pump
4 Freeze flange
5 Thermal shield
6 Coolant pump
7 Radiator
8 Coolant drain tank
9 Fans
10 Fuel drain tank
11 Flush tank
12 Containment
13 Freeze valve



MSRE



Summary of ORNL Experiments

Parameter	Aircraft Reactor Experiment (ARE)	Molten Salt Reactor Experiment (MSRE)
Date of operation	1954	1965-1970
Max. Power (MWth)	2.5	8.0
Max. Temperature (°C)	860	650
Moderator	BeO (solid)	Graphite (solid)
Fuel-Salt composition (%mol)	NaF-ZrF_4-UF_4 (53-41-6)	^7LiF-BeF_2-ZrF_4-UF_4 (65-29.1-5-0.9)
Secondary loop	Na	^7LiF-BeF_2

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Neutronics: Point Kinetics

Assume the flux distribution does not change, only the amplitude: **point kinetics**

Define average neutron **generation time:**

$$\Lambda = \frac{\text{neutron population}}{\text{production rate}}$$

And **reactivity**

$$\rho = \frac{\text{production rate} - \text{loss rate}}{\text{production rate}} = 1 - \frac{1}{k_{\text{eff}}}$$

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Point Kinetics

Now
$$\frac{dn(t)}{dt} = \frac{\rho}{\Lambda} n(t)$$

With obvious solution

$$n(t) = n(0)e^{\frac{\rho}{\Lambda}t}$$

All of this only considers neutrons from fission.

Fortunately, there are **delayed neutrons.**

(Unfortunately, there are **delayed neutrons.)**

Delayed Neutrons

- Fission products are always
  - Radioactive
  - South of the line of stability (too many neutrons)
- Decay towards line of stability by β-decay (electron), followed possibly by emission of a neutron.
- β-decay is slow: ms, s, min, → …
- Emitters are called **precursors**
- Emitted neutrons are **delayed neutrons.**

DN distribution

Table 1 Delayed-neutron data for thermal fission in ^{235}U ([Rose1991])

Group	Decay Constant, λ_k (s^{-1})	Delayed Yield, ν_{dk} (n/fiss.)	Delayed Fraction, β_k
1	0.01334	0.000585	0.000240
2	0.03274	0.003018	0.001238
3	0.1208	0.002881	0.001182
4	0.3028	0.006459	0.002651
5	0.8495	0.002648	0.001087
6	2.853	0.001109	0.000455
Total	-	0.016700	0.006854

$\beta = \sum_{k=1}^{6} \beta_k$ is a crucial parameter in a reactor

Q: How much is it worth?

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Point Kinetics with DN

- Interesting thought: every neutron in a reactor is in a chain that originated in a delayed neutron precursor.
- With DN, the point kinetics equation becomes

$$\frac{dn(t)}{dt} = \frac{\rho - \beta}{\Lambda} n(t) + \lambda C(t)$$

with $C(t)$ the average precursor concentration.

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Precursor Concentration

- Precursors originate in fission, then decay:

$$\frac{dC(t)}{dt} = \frac{\beta}{\Lambda}n(t) - \lambda C(t)$$

- Taking the six precursor groups:

$$\frac{dC_1(t)}{dt} = \frac{\beta_1}{\Lambda}n(t) - \lambda_1 C_1(t)$$

$$\vdots$$

$$\frac{dC_6(t)}{dt} = \frac{\beta_6}{\Lambda}n(t) - \lambda_6 C_6(t)$$

$$\frac{dn(t)}{dt} = \frac{\rho - \beta}{\Lambda}n(t) + \sum_{k=1}^{6} \lambda_k C_k(t)$$

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Point Kinetics in MSR

Recall:

$$\frac{dC_1(t)}{dt} = \frac{\beta_1}{\Lambda}n(t) - \lambda_1 C_1(t)$$
$$\vdots$$
$$\frac{dC_6(t)}{dt} = \frac{\beta_6}{\Lambda}n(t) - \lambda_6 C_6(t)$$

Now (group k=1 only):

u_z is the velocity of the salt flowing in the z direction.

$$\frac{dC_1(z,t)}{dt} = \frac{\partial C_1(z,t)}{\partial t} + u_z \frac{\partial C_1(z,t)}{\partial z}$$
$$= \frac{\beta_1}{\Lambda}n(z,t) - \lambda_1 C_1(z,t)$$

Point Kinetics cont'ed

$$C_k(0,t) = C_k(H, t - \tau_{\text{out}})e^{-\lambda_k \tau_{\text{out}}}$$

$$\tau_{\text{out}} = \tau_{\text{in}} \frac{V_{\text{out}}}{V_{\text{in}}}$$

Bad news:

- Delayed neutron precursors decay outside of core.
  - Reduces beta (β)
  - Affects the controllability of the reactor
  - Activates the outer circuit

MSRE Experience (1969)

Summary of MSRE Nuclear Parameters with ^{235}U and ^{233}U Fuels

Parameter	Units	^{235}U Fuel		^{233}U Fuel	
		Calculated	Measured	Calculated	Measured
Initial critical concentration in salt	g U/liter	33.06[a]	32.85 ± 0.25[a]	15.30[b]	15.15 ± 0.1[b]
Reactivity loss due to circulation of delayed-neutron precursors	% $\delta k/k$	0.222	0.212 ± 0.004	0.093	c
Control-rod worth at initial critical loading[d]	% $\delta k/k$				
1 Rod		2.11	2.26	2.75	2.58
3 Rods, banked		5.46	5.59	7.01	6.9
Temperature coefficient of reactivity at operating loading	$\frac{\delta k/k}{°F}$ $(\times 10^5)$				
Total		-8.1	-7.3 ± 0.2	-8.8	-8.5
Fuel		-4.1	-4.9 ± 2.3	-5.7	e
Concentration coefficient of reactivity	$\frac{\% \delta k/k}{\% \delta c/c}$	0.234	0.223	0.389	0.369

[a] ^{235}U only.
[b] Uranium of the isotopic composition of the material added during the critical experiment (91% ^{233}U).
[c] Measurement obscured by effect of circulating voids.
[d] Normal full travel of rod(s).
[e] Not separately evaluated.

MSRE: Zero-Power Exp.

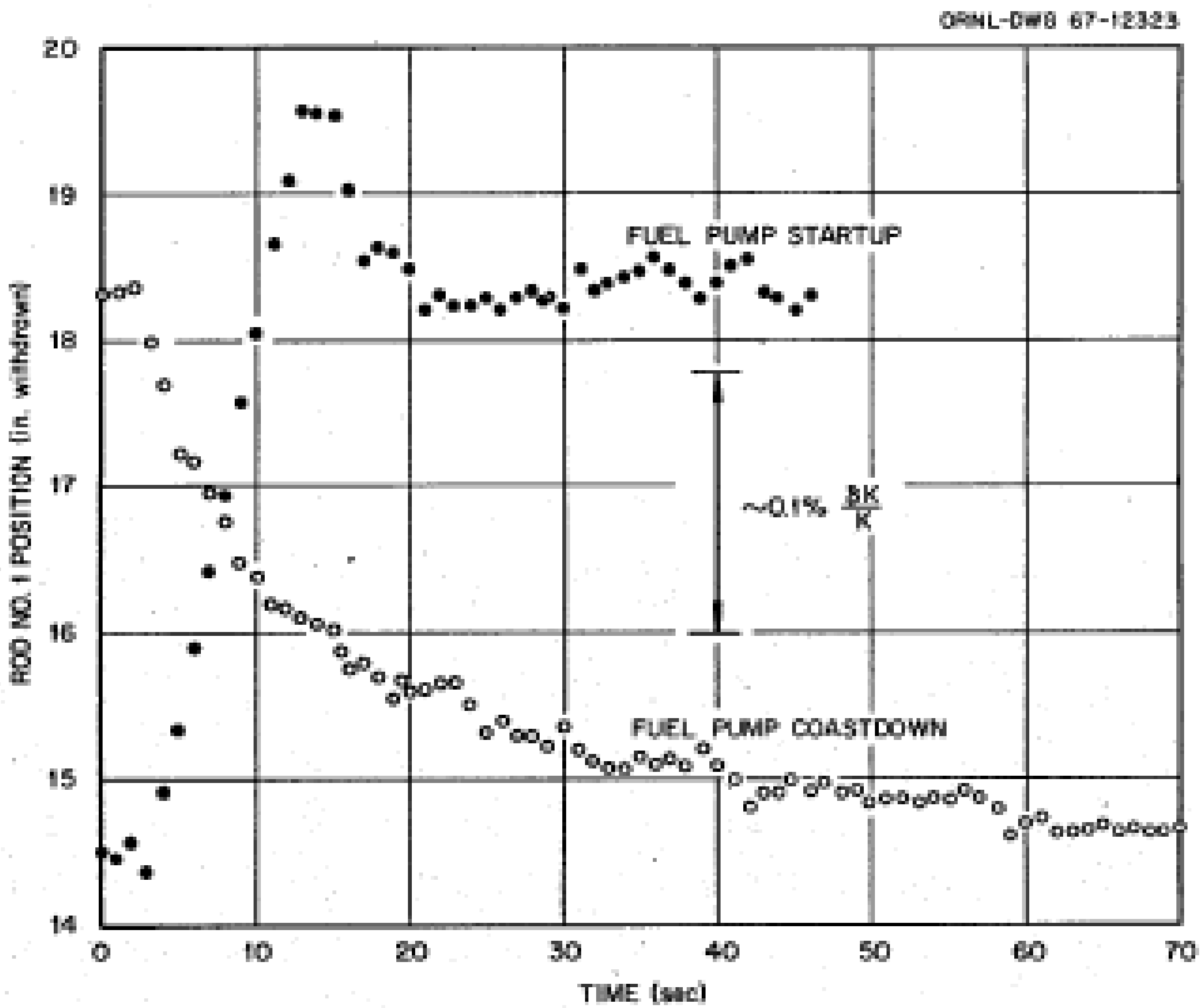
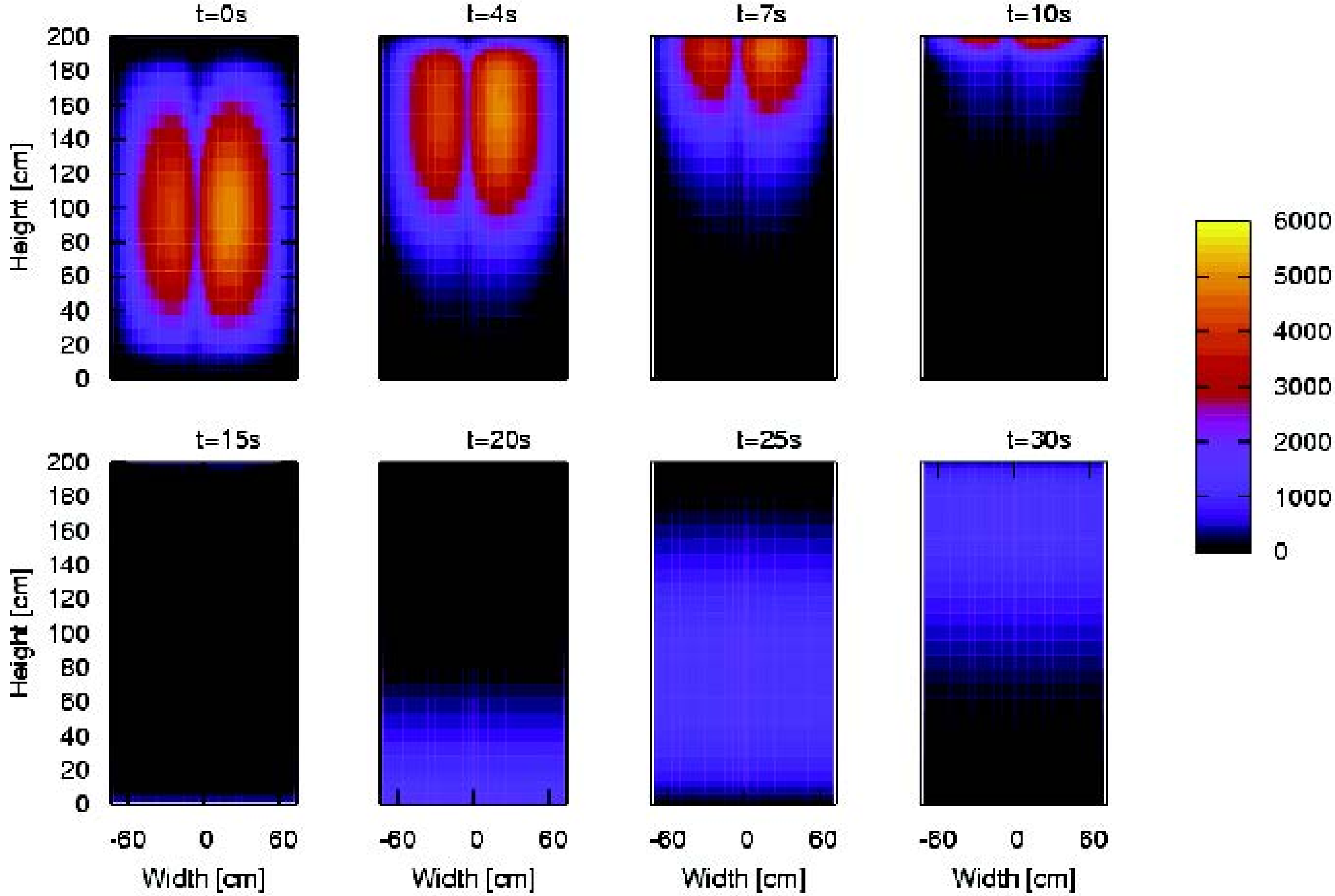


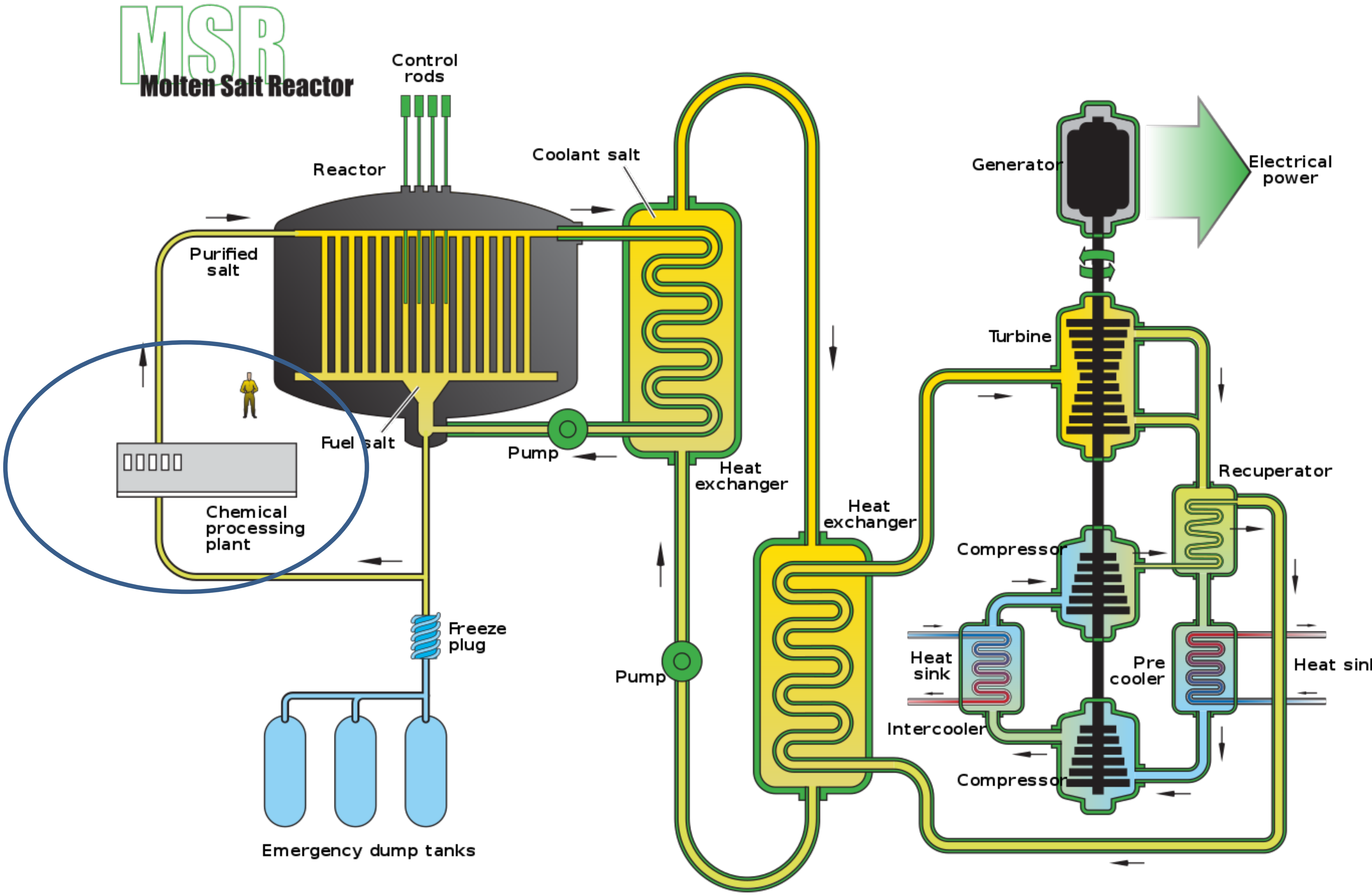
Fig. 24. Control-Rod Response to Fuel-Pump Startup and Coastdown.

MSRE Calculation

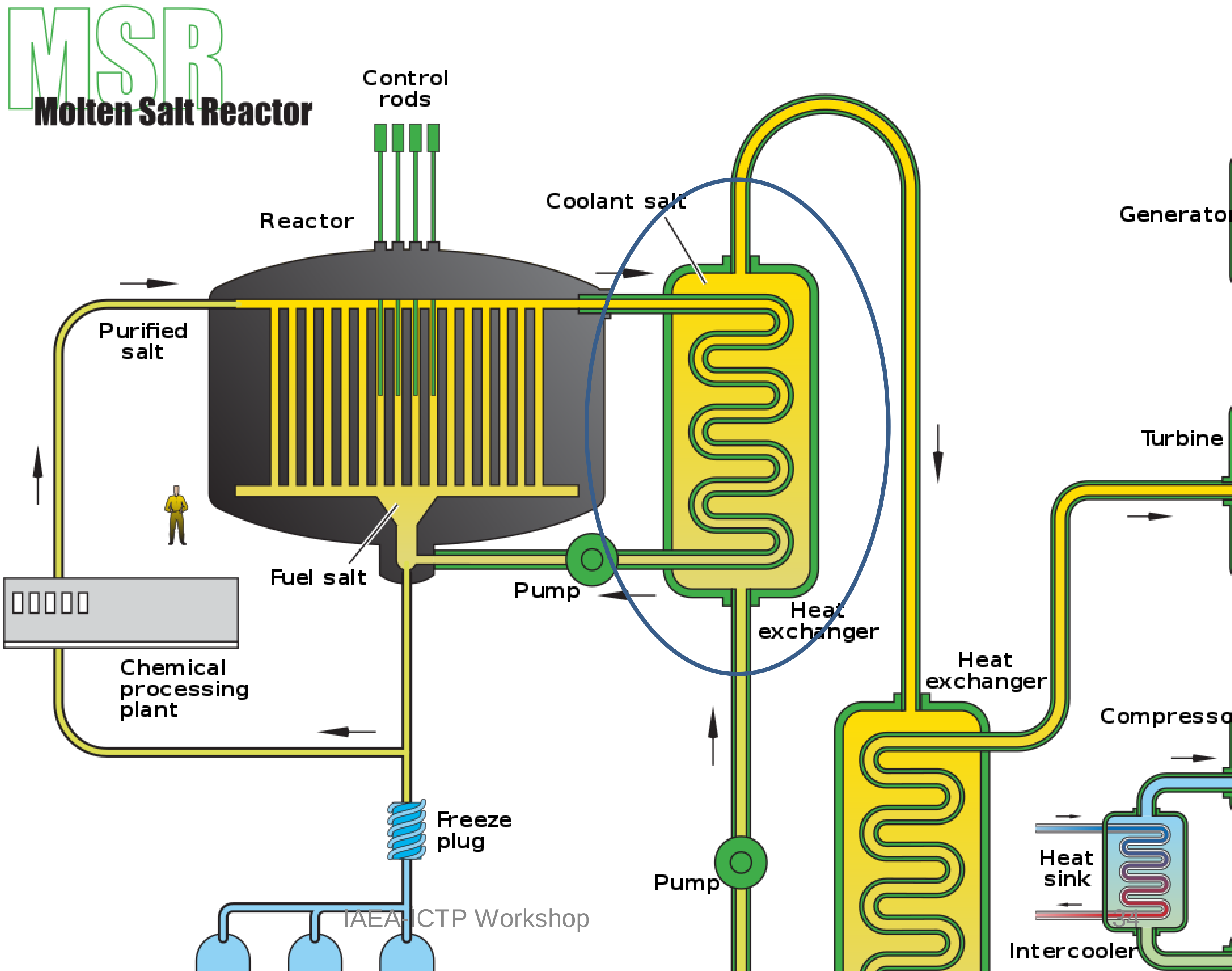


Multiphysics analysis by Danny Lathouwer (TU Delft)
Longest-living precursor group only.

Apply to MSRs



Apply to MSRs



Primary Circuit Outside Core

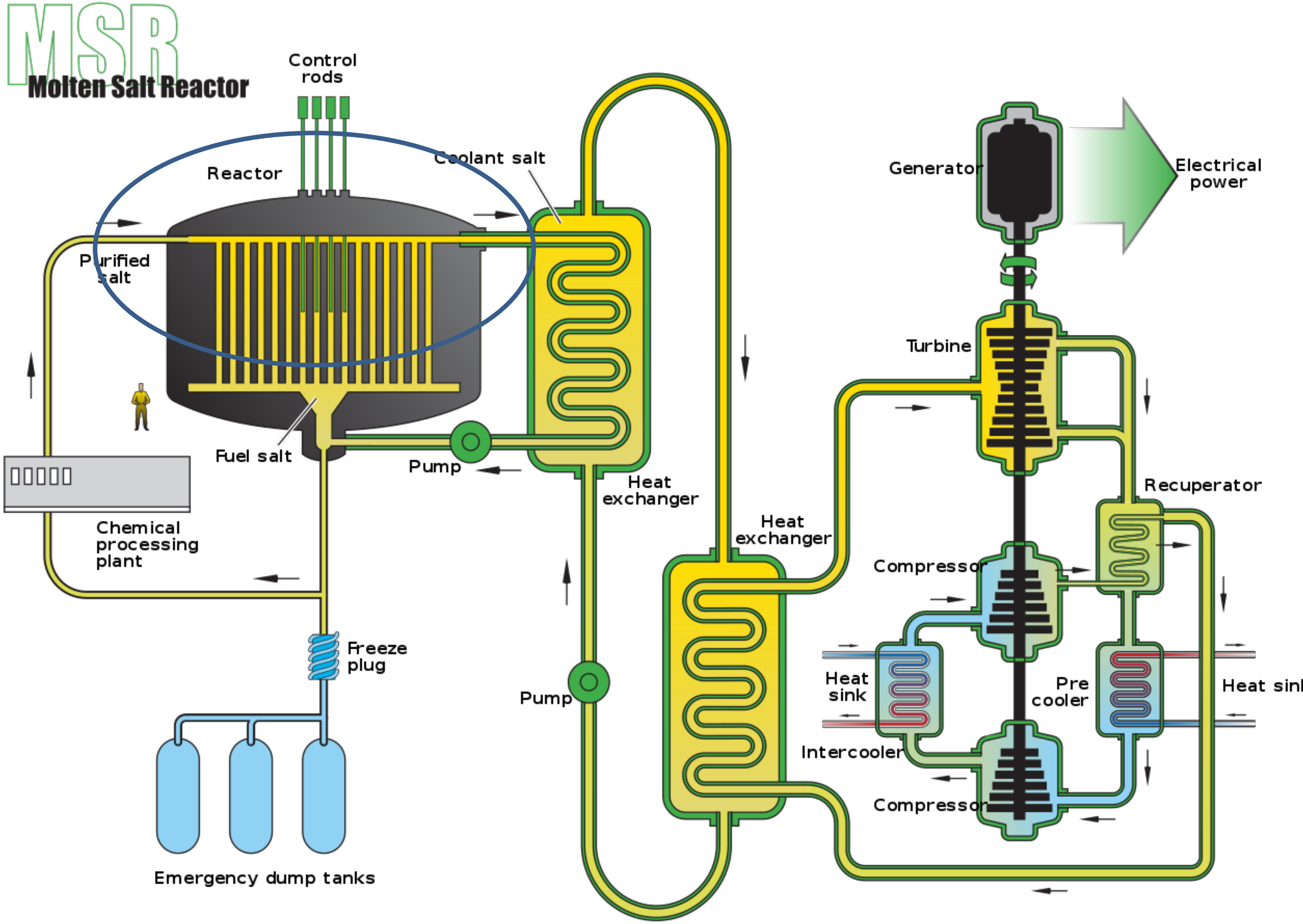
- Good for letting Pa decay
- Ratio: $\mathrm{R} = \dfrac{\text{time in core}}{\text{time out of core}}$ for a given sample of fuel salt.
- Equal to the ratio of volumes: ${}^{V_{\text{in}}}\!/_{V_{\text{out}}}$.
- Small R = good for **Pa decay**.
- Small R = bad for **delayed neutrons**.

$$\text{time in core}(\tau_{\text{in}}) = \frac{\text{height of core } (H)}{\text{liquid velocity } (u)}$$

Chemical Processing Plant

- Remove fission products
  - One of the main design features of original ORNL design.
  - In thorium operation, remove protactinium-233 to let it decay to U-233, avoiding the n-capture.
  - Topping up the fuel, to compensate for burnup.

Apply to MSRs



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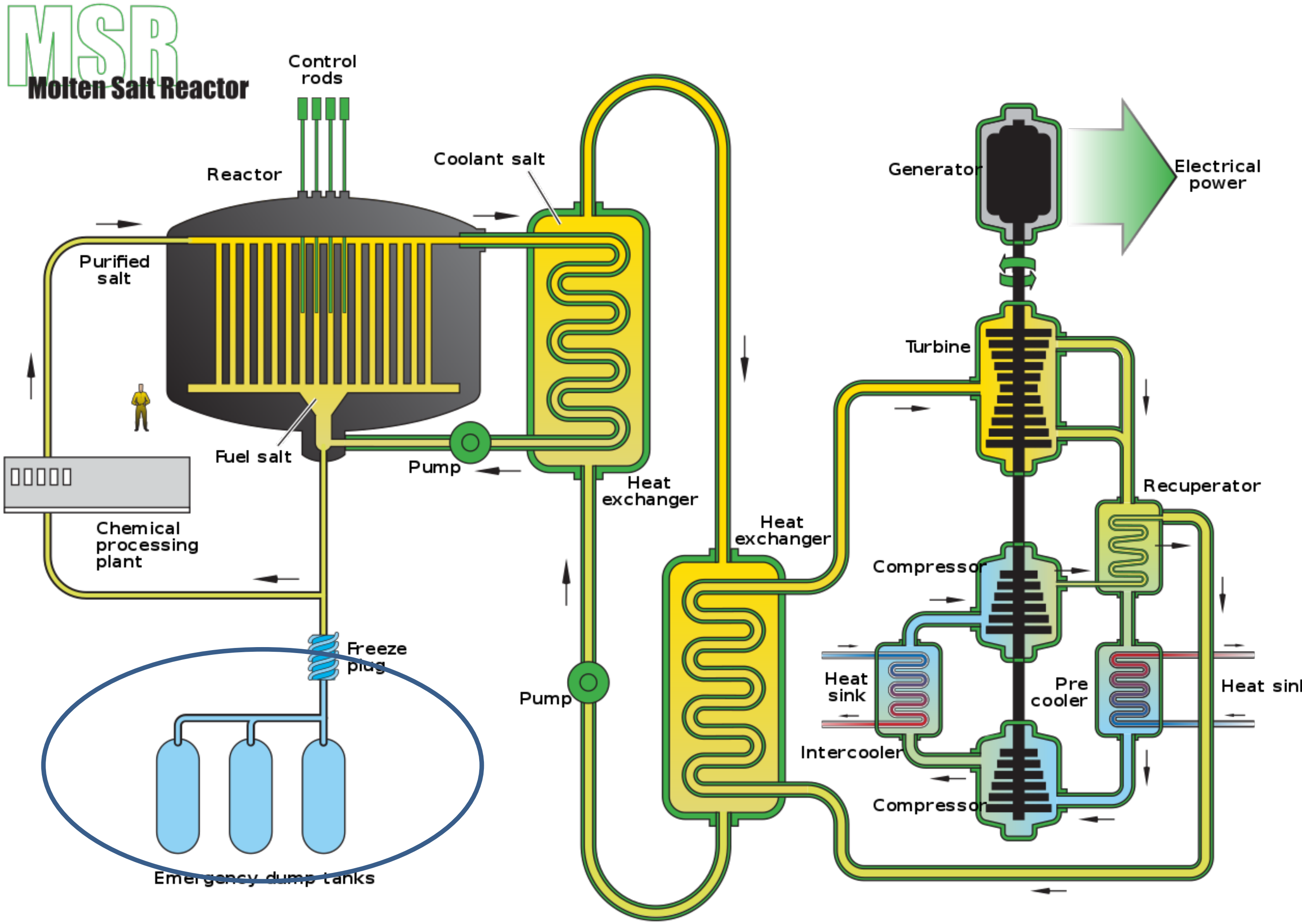
Vessel Head

- Low pressure operation
- “Vent off”, extract fission gases
  - Krypton
  - Xenon, strong n-absorber: no more poisoning out after shutdown, can restart immediately.

Apply to MSRs



Dump Tanks

- Freeze plug: melts when temperature gets too high, fuel is dumped in tanks.
- Still need cooling from decay heat, passive cooling system.
- Worry about flooding.

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Simulating MSRs

- Static (design calculations):
  - Neutronics code; most are satisfactory:
    - MCNP
    - SCALE suite
    - Serpent
    - DRAGON/DONJON
    - ….
  - Depletion code:
    - Serpent
    - TRITON (SCALE)
    - DRAGON
    - ….

Simulating MSRs

- Difficulties:
  - Very strong feedback with T/H.
    - Need iteration to get static solution, e.g. with a code such as RELAP.
    - May need CFD code.
    - Fortunately, only single phase flow.
  - Simulation of delayed neutrons.
  - Effect of Xe removal.
  - Simulation of abnormal conditions
    - Flow blockage
    - Travelling “slugs”, higher/lower density

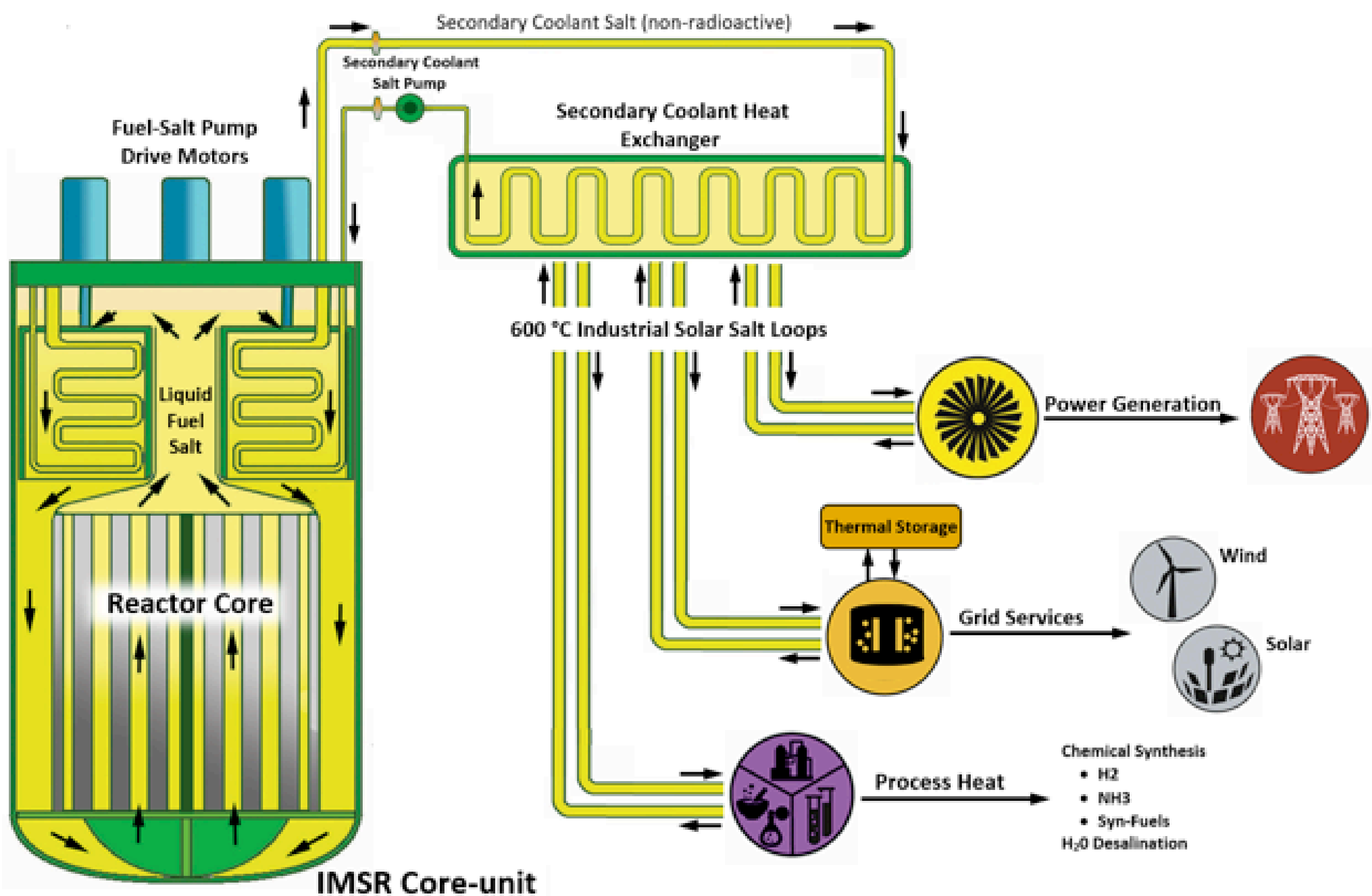
Simulating MSRs

- Much development is being done in this area, notably the Chinese **COUPLE** code: a time-space-dependent coupled neutronic and thermalhydraulics code.
- An important aspect of all these calculations is the determination of sensitivities and uncertainties:
  - E.g. the fuel temperature is negative, but what is the uncertainty? (in other words, how sure are we that it is negative?)
  - Focus has been on S/U due to nuclear data.
  - TSUNAMI, part of SCALE was developed for S/U studies.

IMSR-400 by Terrestrial Energy

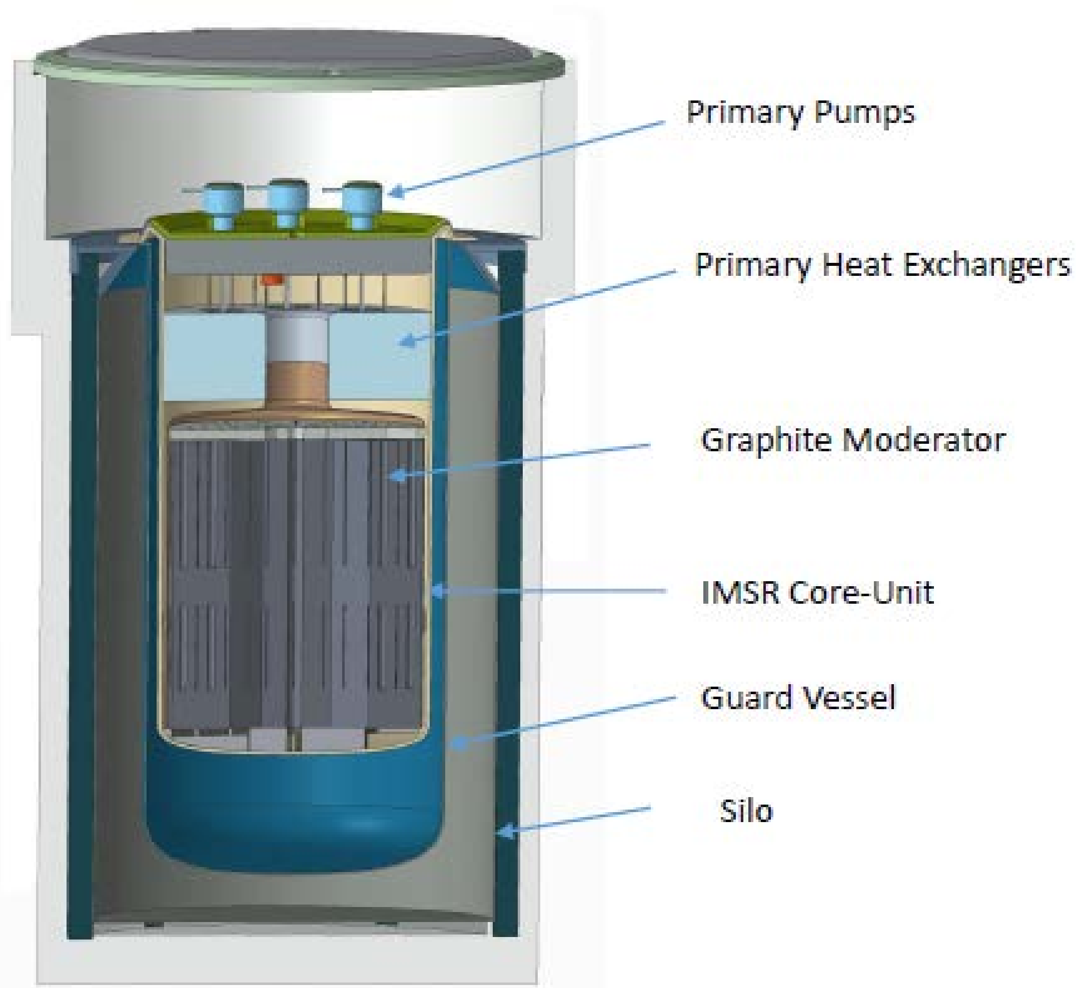
- Based on MSRE experience;
- Modular design (SMR):
  - Two units, one operational, one cooling down
  - Containment is never opened
  - Seven year life-cycle
- Fission gas venting, but
  - No fission product removal
  - No online reprocessing
  - Top up with 20% LEU
- Fuel salt composition proprietary (no Be)

iMSR-400 Design



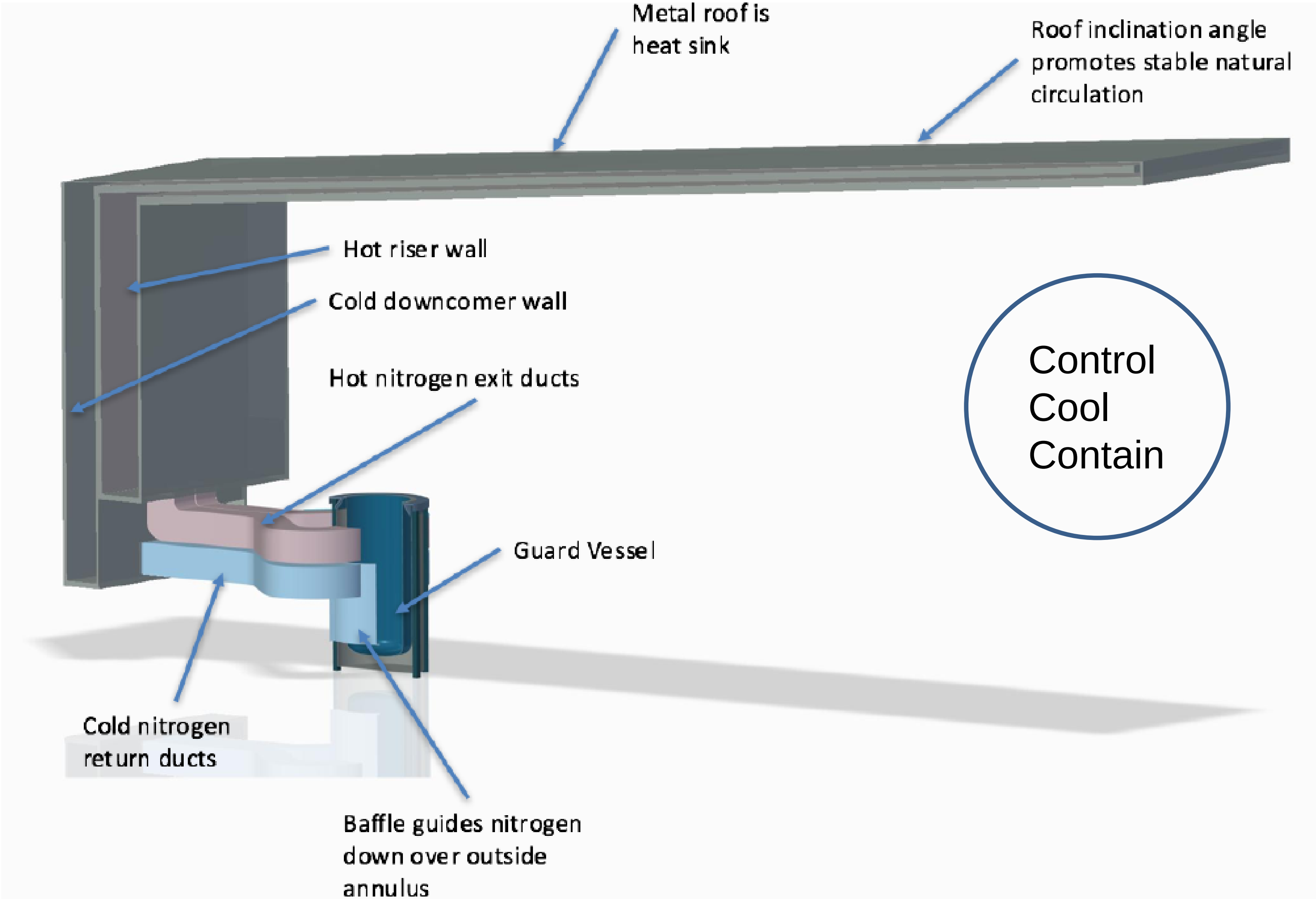
IMSR-400 Core Lay-out

- No dunk-tank!
- Instead always-on passive cooling



Core-unit and guard vessel in one of the two reactor silos

IMSR-400 Passive cooling



Control
Cool
Contain

Last Word: the Regulator

- Each country has its own regulator. Often working with and/or supported by IAEA.
- E.g. Canadian Nuclear Safety Commission
  - Not prescriptive, onus is on vendor
  - Need to prove design is safe
  - Diverse (support) staff, e.g.
    - Rumina Velshi (President)
    - Dumitru Serghiuta
    - Ramzi Jammal
    - Parvaiz Akhtar
    - Nana-Owusua Kwamena
    - Mok Cher Fong

Conclusion

- MSRs have a long history.
- Early designs seem to have been successful.
- Renewed interest in the technology:
  - Private industry
  - Gen IV
  - International collaborations
  - Conservative designs likely to succeed
- MSRs are a safe, reliable and sustainable source of low-carbon electricity.