Dynamic System Simulation of Fissile Solution Systems

Optimized Aqueous Homogeneous Reactor Design for Isotope Production

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June, 2014

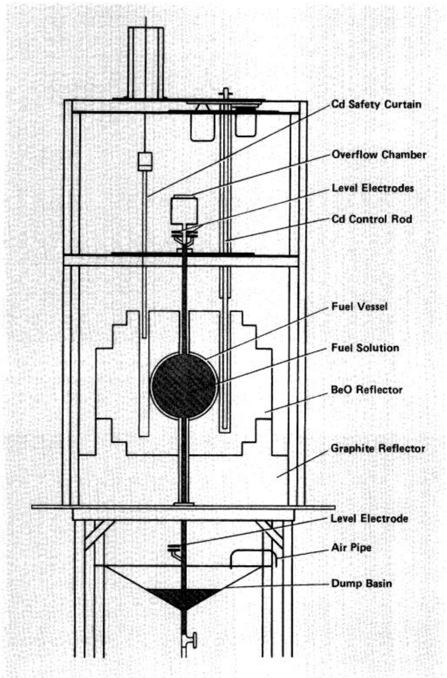
LA-UR-14-22490

Topics & References

- **Topical Outline**
  - Historical Reference
  - Theoretical Studies – System Modeling
  - The Benchmark Systems – SUPO, KEWB, Silene
  - The Way Ahead – Optimized Isotope Production Systems
- **Relevant Publications**
  - A Generic System Model for a Fissile Solution Fueled System, LA-UR-13-22033; Kimpland, Robert H. & Klein, Steven K., July, 2013
  - Neutron Diffusion Model for Prompt Burst Simulation in Fissile Solutions, LA-UR-13-26779; Kimpland, Robert H. & Klein, Steven K., August, 2013
  - A Generic System Model for a Fissile Solution Fueled Assembly – Part II, LA-UR-13-28572; Kimpland, Robert H. & Klein, Steven K., January, 2014
  - Dynamic System Simulation of Fissile Solution Systems, LA-UR-14-22490; Kimpland, Robert H., Klein, Steven K., & Roybal, Marsha M., April, 2014
  - Pumped Fuel Aqueous Homogeneous Reactor, LA-UR-14-23056; Kimpland, Robert H., & Klein, Steven K., April, 2014

The "Water Boilers"

LOPO



LOPO achieved Criticality in May 1944 with Enrico Fermi at Controls; used to determine the critical mass of ^{235}U

HYPO

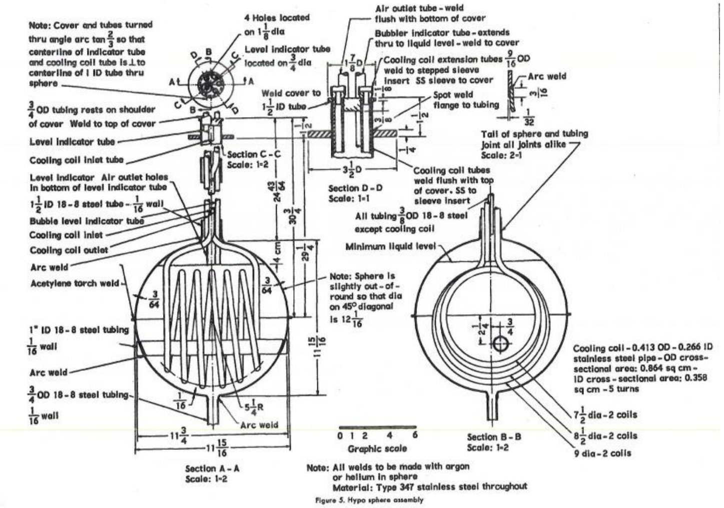


Figure 5. Hypo sphere assembly

Placed into operation in December 1944; many of the key neutron measurements required for design of early atomic weapons were made on HYPO

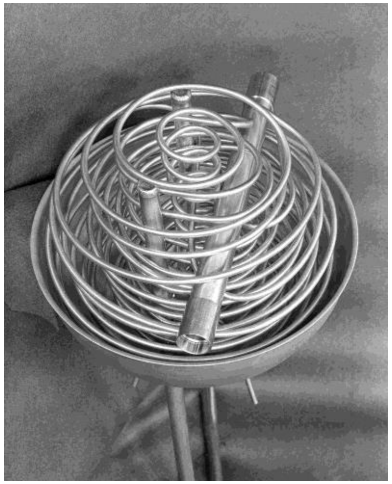
SUPO operated almost daily from 1951 – 1974; supported weapons program particularly in obtaining accurate values of weapon yields

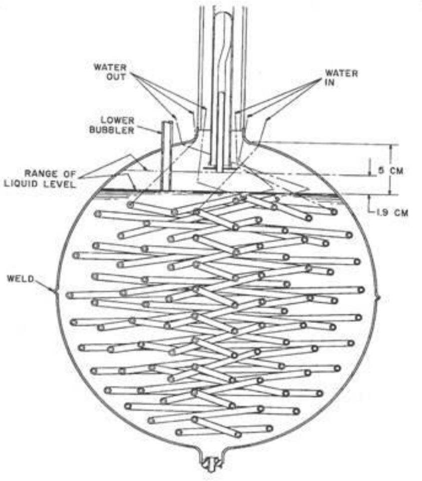
SUPO

SUPO: Prototypical AHR; Steady-State Benchmark

Characteristics

- Operated from 1951-1974
- Accumulated ~600,000 kW-h of operation; typically 25kW (1.7 kW/liter) @ 60° C
- HEU Uranyl Nitrate fuel
- 1 kW – 25 kW (40°C - 60°C)
- Produced ~11 liters/min radiolytic gas @ 25 kW
- Spherical, Graphite Reflected, Cadmium & Boron Control Rods, Actively Cooled



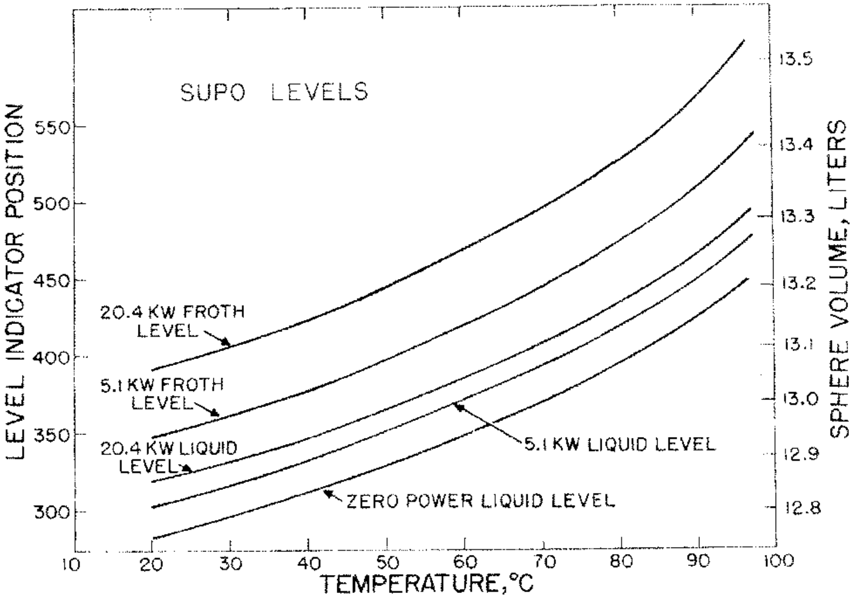


Observations

- Essentially all data on transient behavior of “cold-unsaturated” core; little on steady-state operation of a “hot-saturated” core
- Standard theoretical treatment did not match data

HYPO/SUPO – At least as many questions as answers

- Data
  - On the surface, these curves seem to indicate the bubble void fraction as a function of temperature.
  - However: How many points were used to generate the curves? The level as a function of temperature is purely an analytic function.
  - Nevertheless, these curves provide the only reference to void fraction as a function of temperature!
- Observations
  - "After the HYPO had been run for several hundred kilowatt hours it was observed that its reactivity had increased remarkably."
  - "After some investigation, it was found that the uranyl nitrate was gradually being converted into basic nitrate and that the free nitrate was presumably being carried away by the flushing air." "Chemical tests indicated that about 30% of the nitrogen had disappeared."



Source: L.D.P.King, *International Conference on the Peaceful Uses of Atomic Energy,* "DESIGN AND DESCRIPTION OF WATER BOILER REACTORS, p. 28.

Development of a System Model for AHR

A System Model is a set of coupled nonlinear differential equations that may be solved in time to simulate the dynamics of the overall system.

Components

- Neutron kinetics model that tracks the deposition of fission energy in the fissile solution core. Subsequent changes in fission power due to reactivity feedback are tracked through the reactivity model that itself is coupled to other sub-model parameters
- Radiolytic gas model that separately tracks the generation and transport of gas bubbles in the fissile solution. The key parameter in this model is the void fraction in the fissile solution, which affects both neutronic and thermal-hydraulic behavior of the assembly
- Core thermal model tracks flow of fission energy from solution to primary coolant loop and then to the secondary side of the heat exchanger.
- Plenum model governs flow of mass and energy into and out of the gas plenum, located above the fissile solution core.

Documented in LA-UR-13-22033, LA-UR-13-28572, and LA-UR-14-22490

System Model Input Data

General Physical Parameters
- Universal constants; molar masses
- Isobaric compressibility, thermal conductivity, expansion coefficient, viscosities, diffusivity, specific heat, density of fuel & water
- Material properties

Core Configuration Parameters
- Initial height & volume of fuel
- Cooling structure geometry

Operational Parameters
- Initial fuel & coolant temperatures
- Coolant mass flow
- Plenum inlet pressure
- Maximum reactivity insertion & rate

Core Reactivity Parameters
- Temperature & void feedback coefficients
- Fission fractions by core region
- Importance fractions by core region
- Gas bubble transit time

System Model Versions

Version 1 (LA-UR-13-22033)

- Physical parameters assumed constant over 20ºC - 80ºC operating range
- One cooling structure in form of coils
- Used for “standard” cores such as SUPO, KEWB, or Silene

Version 2 (LA-UR-13-28572)

- Variable physical constants by pressure, temperature, and salt content
- Similar results as Version 1 for same cores but can handle pressurized cores like HRE

Version 3 (LA-UR-14-22490)

- Up to three cooling structures in form of coils, tubes, or annular channels
- Option for sub-critical accelerator-driven neutron source

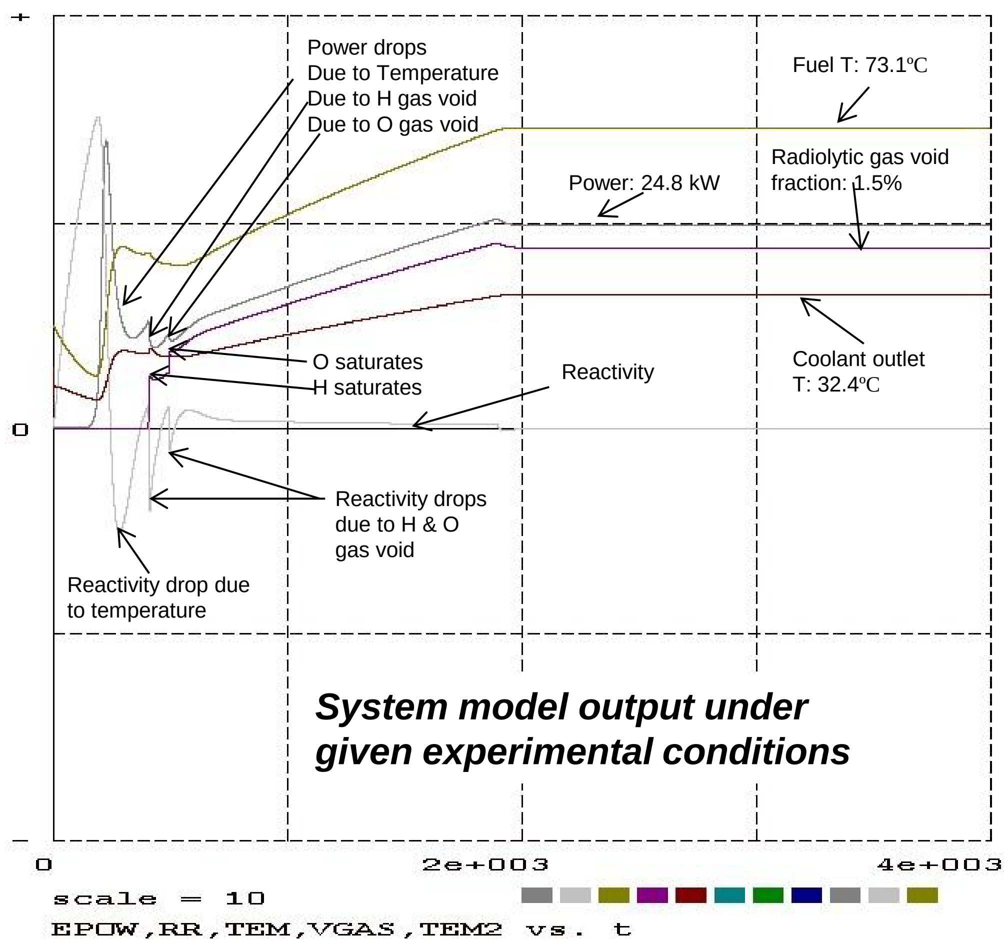
Application of System Model to SUPO Steady-State

^{235}U content of fuel	870 gm
Boron control rod position	52.5%
Sphere cooling water flow	3.43 gal/min
Cooling water inlet temperature	5.0 °C
Cover gas air flow	100 l/min
Excess Reactivity	$1.90

Experimental conditions for SUPO that resulted in 25 kW, fuel temperature of 75ºC and coolant outlet temperature of 35ºC

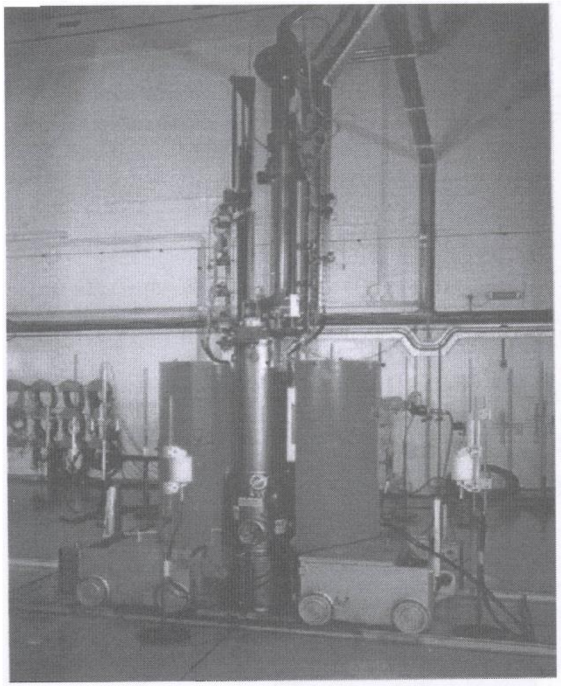
LA-2854, STATUS REPORT ON THE WATER BOILER REACTOR. Merle E. Bunker, February 1963

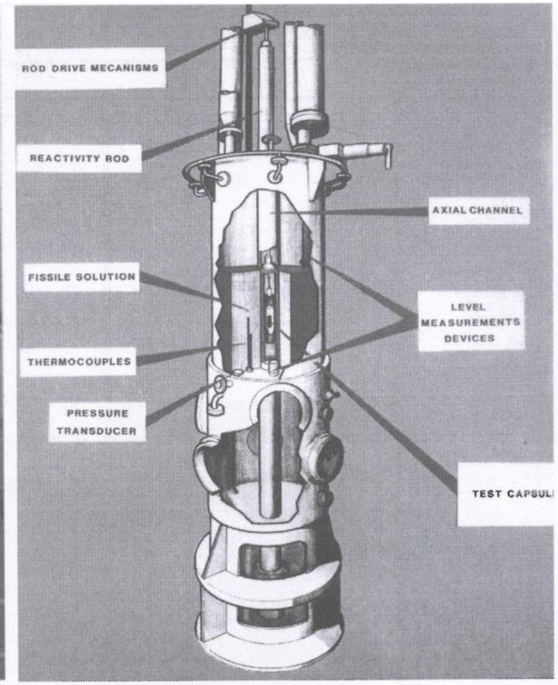
Results show negative reactivity feedback due to temperature and radiolytic gas void



System model output under given experimental conditions

Silene: Benchmark for Pulse Operations





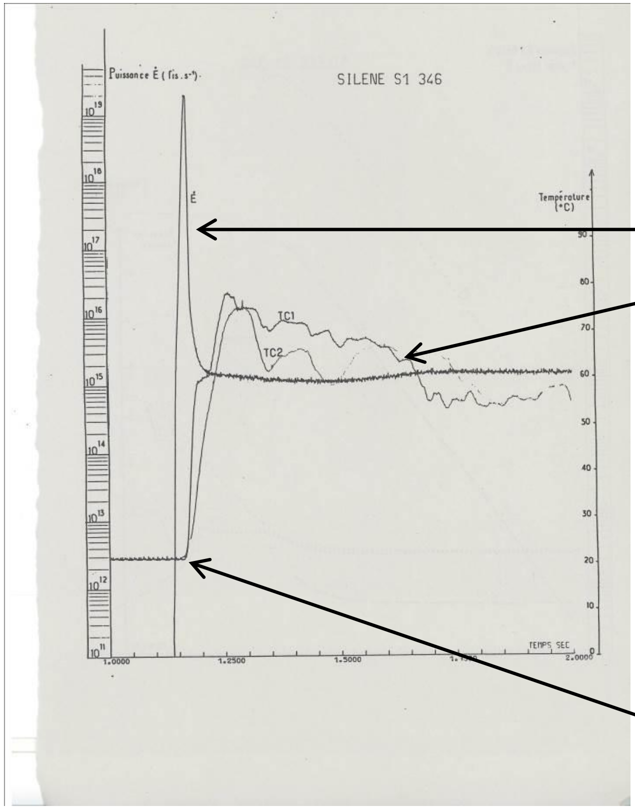
Operations

- Pulse – Δk <u>>></u> β; reactivity insertion rate ~$20.00/sec
- Slow Kinetics – Δk ≤ β; reactivity insertion rate ~$0.03/sec
- Free Evolution – reactivity insertion rate ~$0.20/sec
- Boiling – reactivity insertions Δk ≥ $5.00 with rate ~$0.40/sec

Experimental conditions for Silene and results documented in *CEA IPSN, Report SRSC n° 223-September 1994, Silene Reactor, Results of Selected Typical Experiments, Francis Barbry, 1994*

Silene – Pulse Operations

$\Delta k \underline{>>} \beta$; reactivity insertion rate ~$20.00/sec

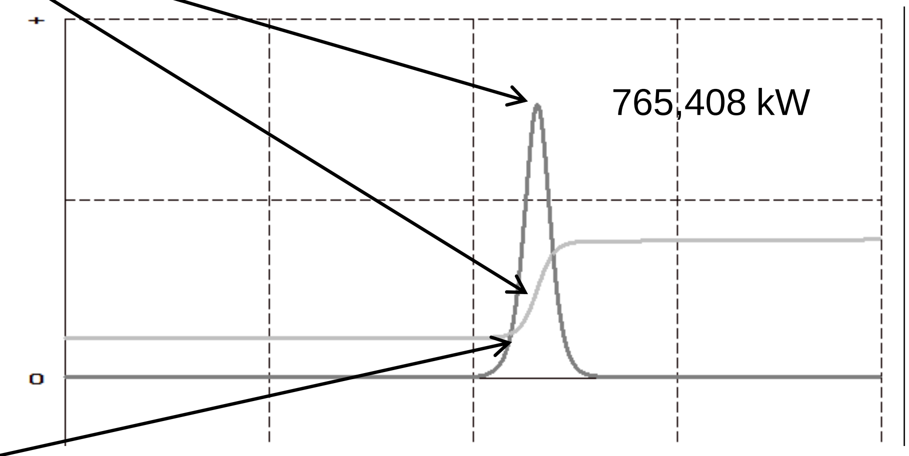


Experimental Trace from S1-364

Experimental Data
- $2.96 step insertion
- 757,576 kW maximum

Power
Temperatures

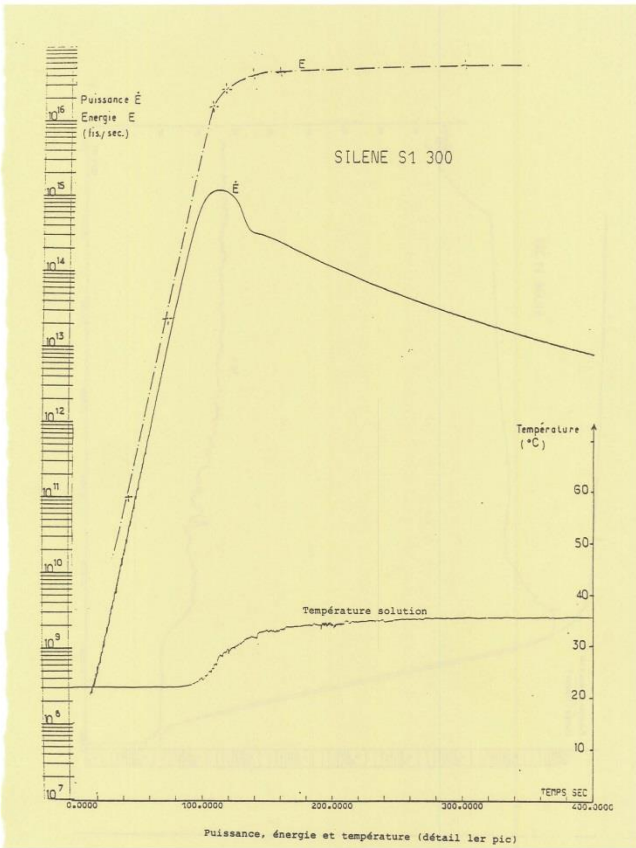
System Model Results
(Normalized Scale)



Power excursion halted by rise in fuel temperature

Silene – Slow Kinetics Operations

$\Delta k \leq \beta$; reactivity insertion ~\$0.03/sec

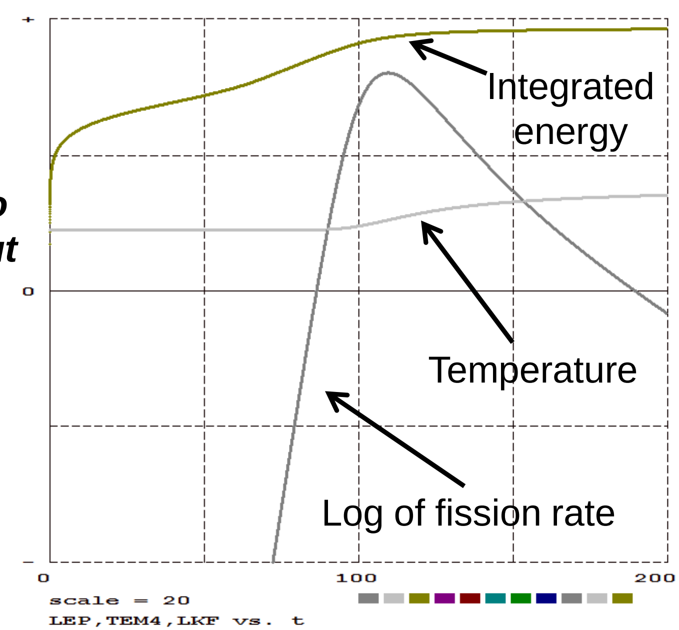


Experimental Trace from S1-300
\$0.51 insertion

Parameter	Experiment	System Model
Peak fission rate	1.3×10^{15}	1.2×10^{15}
Fissions to eq	6.0×10^{16}	7.0×10^{16}
ΔT @ equilibrium	13.7	13.9
Fissions to peak	2.2×10^{16}	1.9×10^{16}

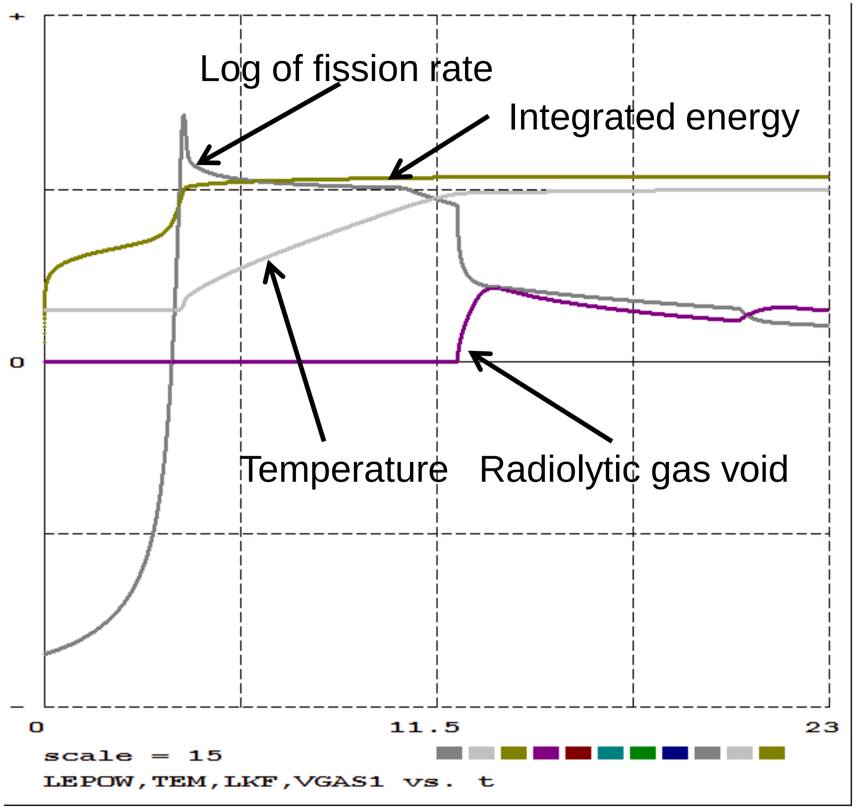
Behavior similar to pulse operation but slower and less energetic

System Model Trace (Normalized Scale)



Silene – Free Evolution Operations

Experiment LE2-362



System Model Trace

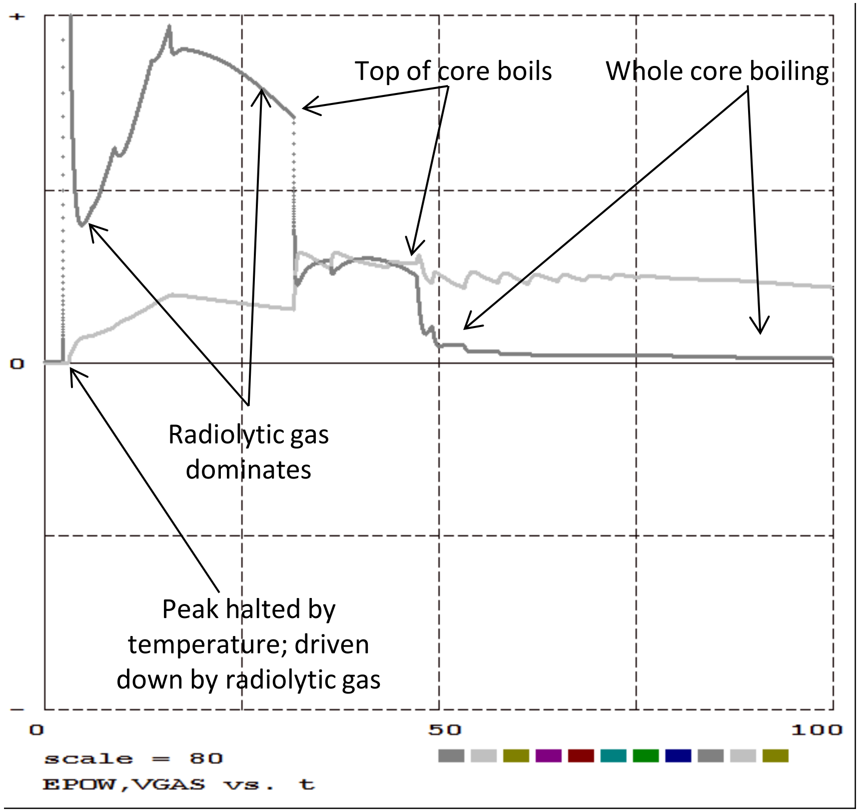
- $2.96 ramp insertion
- $0.28/second

Parameter	Experiment	System Model
Peak fission rate	1.8×10^{17}	2.1×10^{17}
Fissions to eq	2.6×10^{17}	2.9×10^{17}
ΔT @ equilibrium	50	55
Fissions to peak	1.2×10^{16}	1.2×10^{16}

- ***Initial peak halted by fuel temperature feedback***
- ***Subsequent sharp power drops with radiolytic gas void***

Silene – Boiling Operations

Experiment LE1-281



System Model Trace

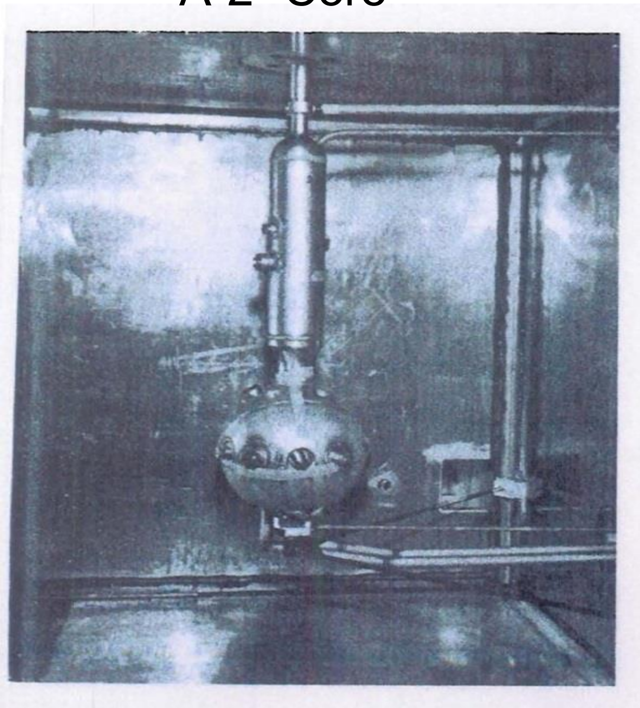
- $7.20 ramp insertion
- $0.45/second

Parameter	Experiment	System Model
Peak fission rate	4.2×10^{17}	3.3×10^{17}
Fissions to peak	1.7×10^{16}	1.6×10^{16}

- ***Early behavior similar to Free Evolution but more energetic***
- ***Power drops dramatically as boiling spreads through core due to steam void***

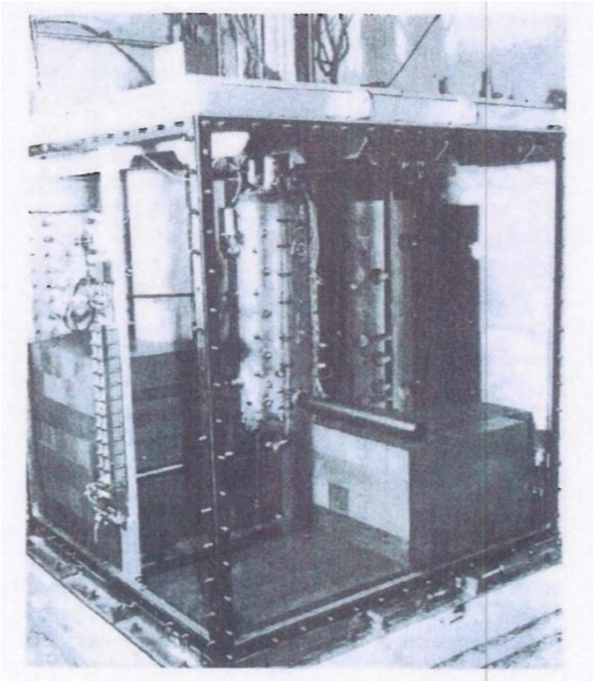
KEWB – Steady-State & Pulse Operations

“A-2” Core



- 12.3” diameter sphere
- 13.7 liters UO_2SO_4
- 106 gU/liter; 93.2% enrichment
- Graphite reflected

“B-5” Core

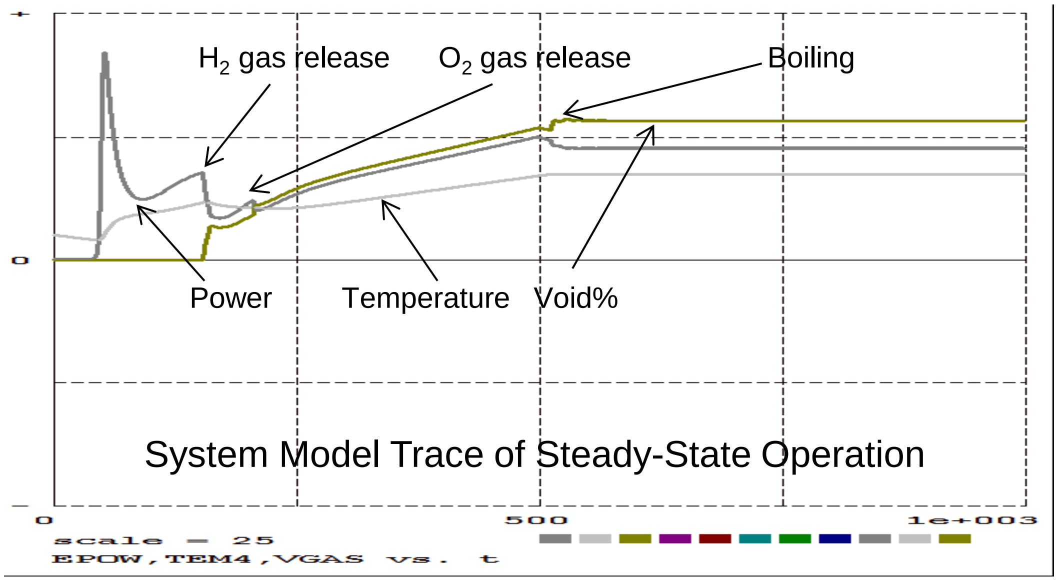


- 12.0” diameter, 36” high cylinder
- 13.7 liters UO_2SO_4
- 203 gU/liter; 93.2% enrichment
- Unreflected

KEWB “A-2”

Operation	$\Delta k_{\$}$	Rate ($/sec)	kW	Temp
Steady-State	5.00	0.01	50	85
	System Model Results		56.78	87.14
Pulse	3.75	Step	6500	N/A
	System Model Results		6470	N/A

- ***Early behavior similar to SUPO Steady-State***
- ***Presence of internal cooling (unlike Silene) allows steady-state operations with boiling***

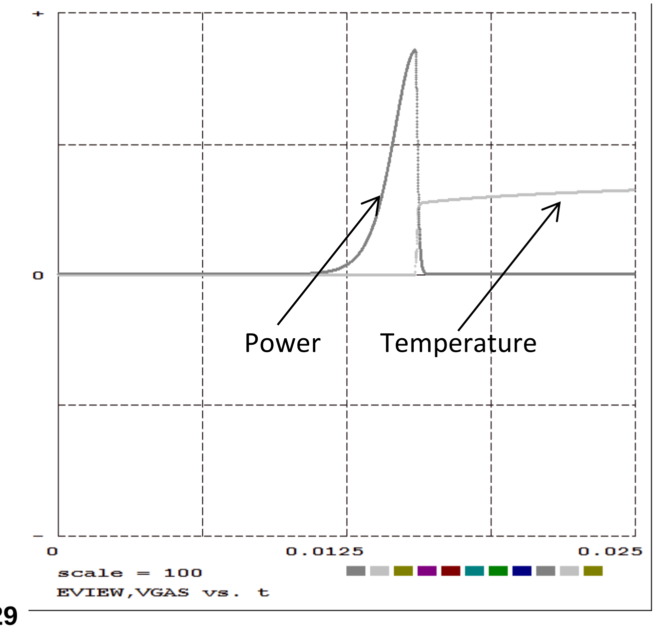


KEWB “B-5” – Pulse Operations

$\Delta k_{\$}$	Peak Power (MW_{exp})	Peak Power (MW_{model})
$3.67	4000	4,072
$3.27	2800	2,941
$2.87	2000	1,680
$2.62	1500	1,149
$2.33	1000	619

System Model Trace of $3.67 Step Insertion

- Asymmetric as compared to Silene
- Sharp downside to peak due to “late” onset of core temperature rise



LA-UR-14-21529

HRE – Pressurized Core Benchmark

HRE-1 Core

Pressurized core critical parameters

- Material constants vary by temperature, pressure, and salt content (thermal conductivity of water, boiling point of water, thermal conductivity of fuel, isobaric compressibility of fuel, expansion coefficient of fuel, kinematic and dynamic viscosities of fuel, specific heat and thermal diffusivity of fuel, density of fuel)

- Operational characteristics vary by temperature and pressure (radiolytic gas and steam bubble transit time, boundary layer thickness, gas saturation threshold concentrations)

- ***HRE-1 operated at 1000 psi nominally producing 1.0 MW***
- ***System model estimates 919 kW at 1000 psi (Boiling at 277ºC)***
- ***Behavior similar to unpressurized cores when critical parameters are considered***

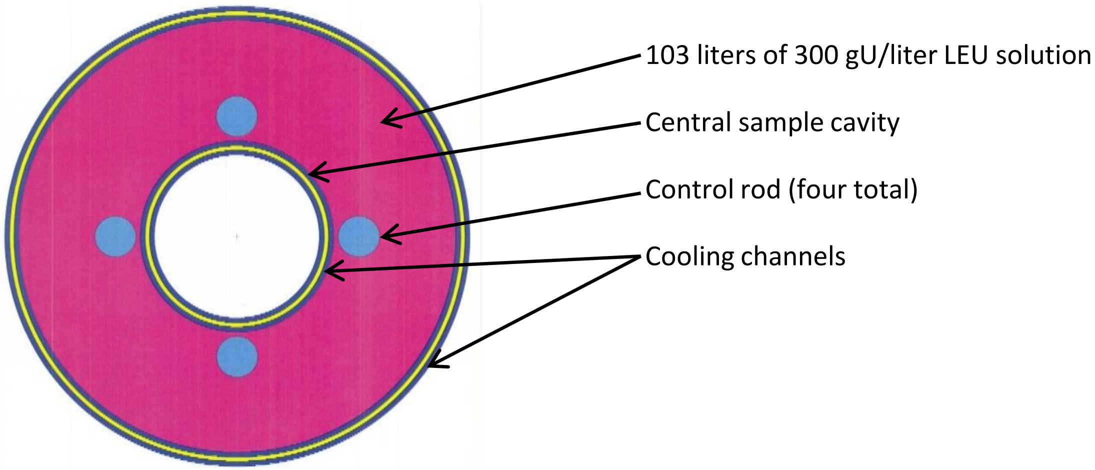
New Design Systems

Super SUPO

- Cylindrical core with SUPO cooling package of 0.25” coils totaling 5% of core volume
- 250 liters of 115 gU/liter, 19.75% enriched (LEU) fuel

Core	Δk($)	kW	Temp (ºC)	Void (%)
SUPO Experiment	1.90	25.00	60.80	Not Reported
SUPO Model	1.90	25.00	64.63	2.21
KEWB “A-2” Experiment	5.00	50.00	85.00	Not Reported
KEWB “A-2” Model	5.00	49.59	84.50	3.99
Super SUPO Basic	1.90	532.89	66.14	3.56
Super SUPO Maximum	4.15	955.10	98.02	8.52

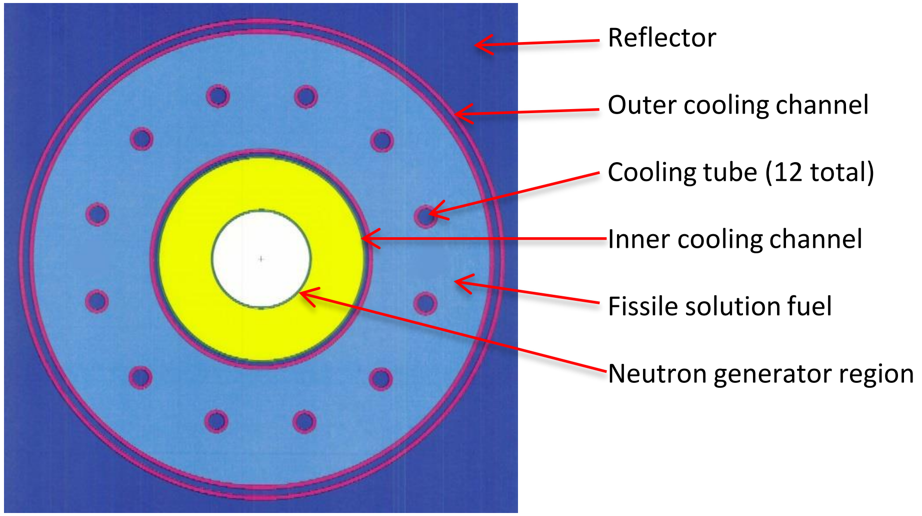
Annular Core AHR



Results for $5.00 Insertion

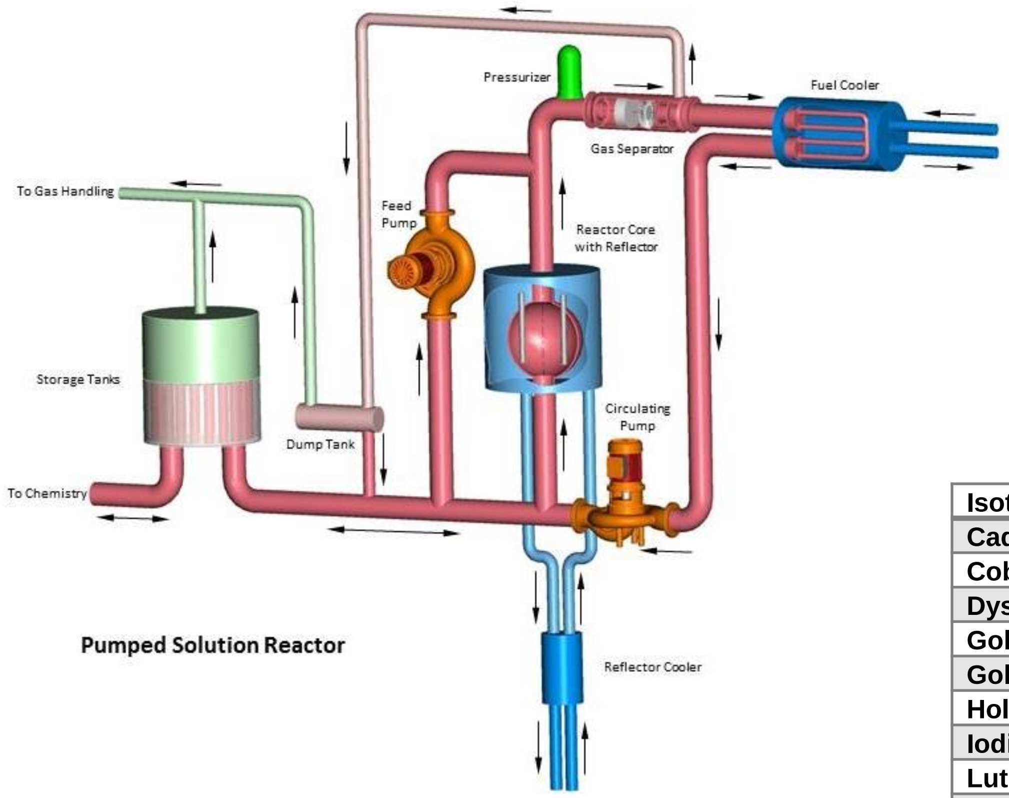
Parameter	Performance
Steady-State Power (kW)	111.20
Fuel Temperature (°C)	82.82
Gas Void (%)	2.08
Step Insertion Power (kW)	217,745

Sub-Critical Accelerator-Driven System



Parameter	Results
Steady-State Power (kW)	71.31
Fuel Temperature (ºC)	60.18
Gas Void (%)	0.81
Inner Channel Outlet Temperature (ºC)	22.78
Cooling Tubes Outlet Temperature (ºC)	22.56
Outlet Channel Outlet Temperature (ºC)	21.40

Pumped Fuel AHR – 1 MW to 3 MW+ System



Fission Products: 1MW, 5 days

Isotope	Symbol	Curies
Cerium-144	Ce-144	9469
Cesium-137	Cs-137	16.63
Iodine-131	I-131	8260
Krypton-85	Kr-85	2.114
Molybdenum-99	Mo-99	37061
Strontium-89	Sr-89	2670
Strontium-90	Sr-90	16.22
Xenon-133	Xe-133	21754
Yttrium-90	Y-90	0.31

Target Products: 1MW, 5 days

Isotope	Symbol	Curies/target gram
Cadmium-109	Cd-109	0.086
Cobalt-60	Co-60	0.200
Dysprosium-166	Dy-166	0.002
Gold-198	Au-198	35.6
Gold-199	Au-199	0.50
Holmium-166	Ho-166	69.8
Iodine-125	I-125	72.8
Lutetium-177	Lu-177	59.2
Palladium-103	Pd-103	5.8
Rhenium-186	Pe-186	33.0
Samarium-153	Sa-153	96.8
Selenium-75	Se-75	1.66
Tellurium-123m	Te-123m	0.195
Tin-117m	Sn-117m	0.50

Aqueous Homogeneous Reactors (AHR) - Summary

- **Large design space**
  - Solution fuels may be virtually any aqueous solution of uranium in concentrations ranging up to point of precipitation
  - Vessel configuration largely dependent on application; neutronics can accommodate a wide range of geometries
  - Operating power ultimately determined by the ability of the cooling system to remove fission generated heat in the solution fuel
- **Operating characteristics**
  - Can operate in modes ranging from steady-state to prompt critical excursions depending on amount of excess reactivity induced and its rate of insertion
  - Dominated by very large negative feedback due to temperature rise and presence of void due to radiolytic gas generation and transport
  - Any bounded reactivity excursion (negative or positive) will result in a bounded response with dynamic behavior reaching a new steady-state position.
- **Optimized Systems for Isotope Production**
  - “Conventional” SUPO derivative capable of 400 kW – 500 kW
  - Pumped AHR capable of operating above 1 MW