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Author(s): Klein, Steven K.
Kimpland, Robert H.

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Discussion Regarding Aqueous Homogeneous Reactor (AHR) Benchmarks

Robert Kimpland & Steven Klein

*Los Alamos National Laboratory
Advanced Nuclear Technology Group (NEN-2)*

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Introduction

“Characteristics of Fissile Solution Systems”, LA-UR-14-21513, published by the same authors, detailed the behavior of the subject systems that distinguishes them from those employing solid fuels. This Report, a companion to the presentation by the same name published by the authors as LA-UR-14-21529, extends that discussion to illustrate this behavior with reference to the operation of historic aqueous homogeneous reactors (AHR).

In this report the term “benchmark” refers to those systems for which it is judged that sufficient experimental data exists to serve as a reference for the behavior of the class of fissile solution systems called AHR. As has been discussed in the previous report, AHR exhibit a range of modes of operation generally determined by the amount of excess reactivity inserted into the system from a steady-state condition, such as sub-critical startup, and the rate of that insertion. The amount of reactivity inserted may fall in the range of $\$0.00 < \text{Reactivity} \leq \1.00 , in the usual units of “dollars” and “cents” or $> \$1.00$. The initial region is called “delayed critical” while the latter is referenced as “super prompt critical”. Excess reactivity may be inserted in rates ranging from slow ramp to step, which drives system dynamics; however, regardless of either of these factors, amplitude or rate of insertion, the ultimate result will be steady-state operation.

Three AHR benchmarks will be discussed. For steady-state operation SUPO (Super Power), which operated for 23 years at Los Alamos National Laboratory (LANL), is chosen. Pulse operations will reference Silene, which operated for many years at the Centre d’Etudes de Valduc, France. Operational characteristics peculiar to pressurized cores will be discussed with reference to the Homogeneous Reactor Experiment (HRE-1) from Oak Ridge National Laboratory (ORNL). In addition, much relevant data is available from the Kinetics Experiments on Water Boilers (KEWB) family of AHR operated by Atomics International, so appeal is made to this information to illustrate behavior.

As was also discussed in the prior report, the principle use of all these AHR was as a source of thermal neutrons for a variety of experiments. The focus of these experiments was generally on the effects of the neutrons on various samples rather than the operation of the AHR itself. This situation resulted in little information being available regarding core physics. To remedy this situation, LANL embarked on a program to develop theoretical models, using Dynamic System Simulation (DSS) techniques, to aid in

understanding AHR behavior. In this report experimental data will often be accompanied by system model results. This comparison will accomplish two goals: first, demonstrate the close agreement between DSS model results and experimental data; second, assist in illustrating AHR behavior.

The “Water Boilers”

The series of three AHR were constructed and operated at LANL during the period of 1944 to 1974 were generally known as the “water boilers”. These reactors were labeled with this moniker due to the observed vigorous bubbling on the solution surface, reminiscent of a boiling pot of water, when operating at power. Even though this term stuck and was used for many years in referring to this class of solution fueled reactors, it was quickly understood that unless sufficient excess reactivity was purposefully introduced into the system, the actual boiling point of water was never approached; the dynamic foaming, bubbling, and even sloshing about of the liquid fuel surface was the result of the transport of radiolytic gas, principally molecular hydrogen and oxygen, from the core.

These three AHR were named LOPO (Low Power), HYPO (High Power), and SUPO. All three were configured as 12 inch diameter spheres. LOPO’s fuel was uranyl sulfate while the other two used uranyl nitrate since uranium metal dissolves better in nitric acid. LOPO and HYPO’s fuel was about 14% enriched with ^{235}U , but by the time SUPO was built a sufficient amount of highly enriched uranium was available so that 88% enriched fuel was used. LOPO first achieved criticality in May of 1944 with the Nobel Prize winning physicist Enrico Fermi at the controls and contributed to the Manhattan Project by allowing the determination of the critical mass of ^{235}U . HYPO was used from the beginning of operation in December 1944 until decommissioned in April 1949 after amassing about 14,000 kilowatt hours. During that time its principal use was to examine key design parameters of early atomic weapons. Operation of SUPO started in 1951 and it operated on a near daily basis until decommissioned in 1974. As with its predecessors, most measurements were directed at obtaining nuclear weapon design data including specific campaigns to assist in the determination of accurate values of weapon yields. The history of these three machines is documented in the landmark papers of H. L. Anderson¹ and L. D. P. King². Figure 1 is a picture and a schematic of SUPO. The 20 feet of 0.25 inch o.d. stainless steel cooling coils can be seen in both representations. The photograph is of the inverted core with the plenum and control rods seen at the bottom of the photograph. The horizontal structure seen in the photograph is a sample tube located at approximately the highest flux position of the reactor. The graphite reflector is not shown.

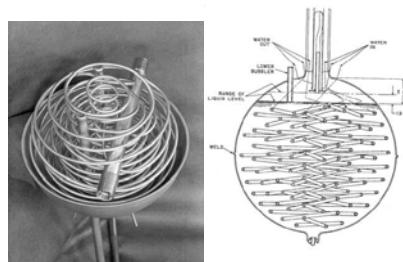


Figure 1: SUPO

¹ H. L. Anderson et. al, LA-394, “HIGH POWER WATER BOILER”, September 19, 1945

² L. D. P. King, LA-1301, THE LOS ALAMOS HOMOGENEOUS REACTOR, SUPO MODEL, October, 1951

During 23 years of operation, SUPO amassed approximately 600,000 kilowatt hours. SUPO operated at critical with a fission power anywhere in the range of 1 kW to 25 kW, exhibiting a fuel temperature of 40°C - 60°C (illustrating the mythology of the “water boiler” moniker). On a few occasions SUPO was pushed to find the threshold of boiling, which occurred at approximately 35 kW at the 0.8 atm of Los Alamos, but the dynamics of the system drove the experimenters to place an operational limitation at 25 kW. SUPO is considered the benchmark for steady-state AHR operation.

Since the purpose of SUPO was as a thermal neutron generator, essentially all data was on the transient behavior of a cold core that was unsaturated with radiolytic gas. Very few experiments extended past a few thousand seconds so the long term behavior of a hot core saturated with gas at steady-state was not studied. Essentially all data relevant to steady-state operation is anecdotal.

King³ reported that “after the HYPO had been run for several hundred kilowatt hours it was observed that its reactivity had increased remarkably” and that, “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.”

Regarding radiolytic gas generation and the effect on fuel density King stated further that SUPO generated approximately 11 liters of radiolytic gas at 25 kW and included the graph shown in Figure 2.

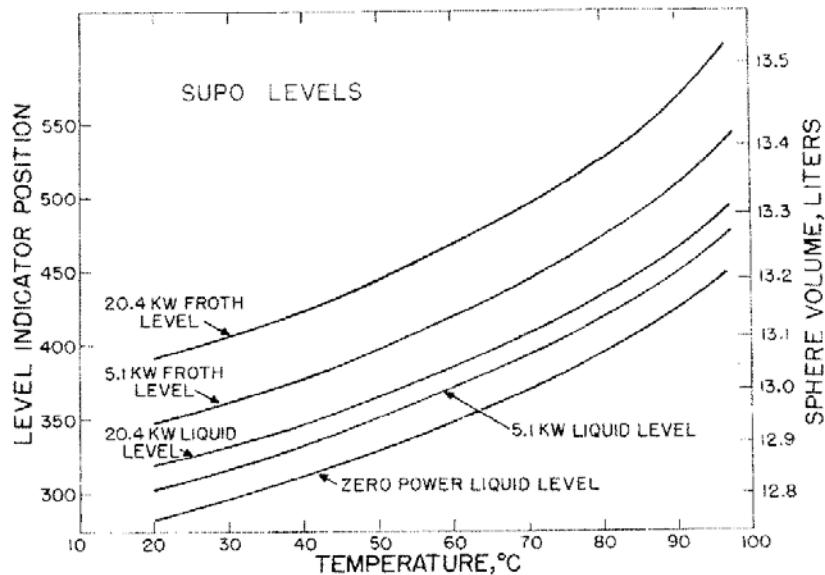


Figure 2: Fuel Height and Volume as a Function of Operating Temperature in SUPO

These curves present the increase in fuel and froth level as judged through an optical viewing sight during operations at various power levels and fuel temperatures. Since the fuel density does not dramatically change due to temperature over this range, the changes may be taken to be due to

³ L.D.P.King, *International Conference on the Peaceful Uses of Atomic Energy*, “DESIGN AND DESCRIPTION OF WATER BOILER REACTORS

radiolytic gas void. These curves provide the only data available on void fraction as a function of temperature and clearly they are drawn curves representing a purely analytic function. No data points were given, either on the graph or in the accompanying logbook.

Development of a System Model for AHR

The paucity of experimental data on the operation of the water boilers, coupled with the lack of an operational AHR specifically designed to explore core physics, leaves system modeling as the only path to a deeper understanding of the operation of these machines. Such theoretical treatment must predict the time evolution of fission power, core temperature, and radiolytic gas void in the core and do so while maintaining energy balance. Dynamic System Simulation (DSS) techniques are particularly suited to this sort of problem. DSS allow the development of a system model as a set of coupled nonlinear differential or difference equations that may be solved in time to simulate system dynamics.

Such a system model has been developed by the authors for an AHR and documented as LA-UR-13-22033⁴ and LA-UR-13-28572⁵. This model has four sub-models or components. First is a neutron kinetics model that tracks the deposition of fission energy in the solution core. Changes in fission power with time due to reactivity feedback are tracked through a reactivity model that is itself coupled to other sub-models dealing with core thermal hydraulics, radiolytic gas generation and transport, and a plenum sub-model governing the pressure on the core.

Application of the System Model to SUPO

Bunker⁶ detailed the operational conditions shown in Table 1 for SUPO.

Table 1: Experimental Conditions for 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 liters/min
Excess reactivity	\$1.90

Using the techniques described in Reference 5, the SUPO System Model may be run and the results compared to experimental data. Bunker documented that with the experimental conditions presented in Table 1 SUPO operated for 4,000 seconds with a steady-state power of 25 kW, fuel temperature of 75°C and a coolant water outlet temperature of 35°C. Figure 3 presents the graphical trace of the output from the system model under these conditions. Note that the power (24.8 kW), fuel temperature (73.1°C), and coolant outlet temperature (32.4°C) compare favorably with experimental results.

⁴ Kimpland, Robert H. & Klein, Steven K., "A Generic System Model for a Fissile Solution Fueled Assembly", LA-UR-13-22033, 2013

⁵ Kimpland, Robert H. & Klein, Steven K., "A Generic System Model for A Fissile Solution Fueled Assembly – Part II", LA-UR-13-28572, 2013

⁶ Merle E. Bunker, LA-2854, STATUS REPORT ON THE WATER BOILER REACTOR., February 1963

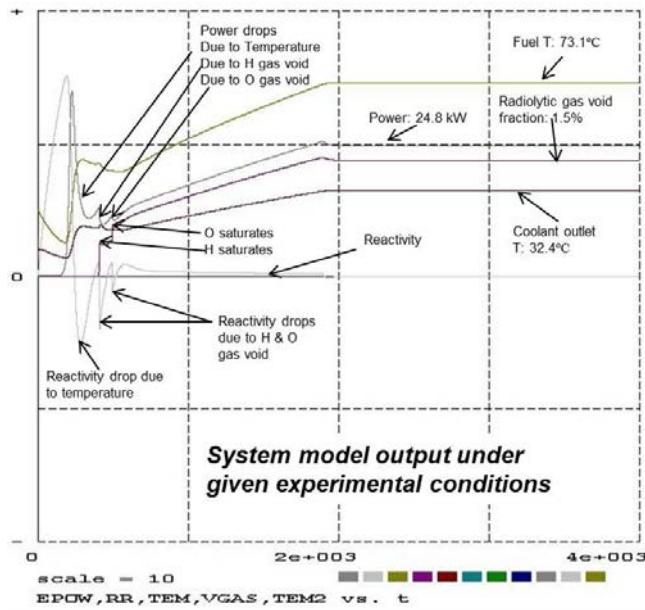


Figure 3: System Model Output for SUPO Steady-State

Figure 3 illustrates that SUPO exhibited the general characteristics of a fissile solution system. The power trace has an initial peak that rises until the onset of fuel temperature increase and then drops sharply, corresponding to the temperature rise. Core reactivity mirrors this behavior. Subsequently, core dynamics exhibits successive decreases due to the void generated, first as hydrogen gas saturates, and then oxygen gas. This behavior shows the negative reactivity effects of temperature and void. Note that these decreases are superimposed on the gradual ramp insertion of the \$1.90 excess reactivity and are reflected in the reactivity trace. At approximately 1,900 seconds the 0.01 \$/second reactivity insertion reaches its maximum value. Thereafter the core establishes a steady-state condition where power, temperatures, and gas void equilibrate.

Kasten⁷ reported on the response of SUPO to rapid reactivity insertions in the delayed critical range. Table 2 is a summary of one of those experiments.

Table 2: Experimental Conditions for SUPO Steady-State

Reactivity insertion rate	1.20 \$/sec
Point of time of reactivity insertion	3,000 sec
Maximum reactivity inserted	\$0.48
Coolant inlet temperature	25.2°C
Coolant mass flow rate	0.1733 kg/sec
Initial power	\$0.10

The reported results of this experiment were power of 31 kW; fuel temperature of 30.3°C, and a coolant outlet temperature of 26.5°C.

⁷ Paul R. Kasten, Nuclear Engineering, C.E.P. Symposium Series, REACTOR DYNAMICS OF THE LOS ALAMOS WATER BOILER, 1954.

Figure 4 is a trace of the system model results for this experiment.

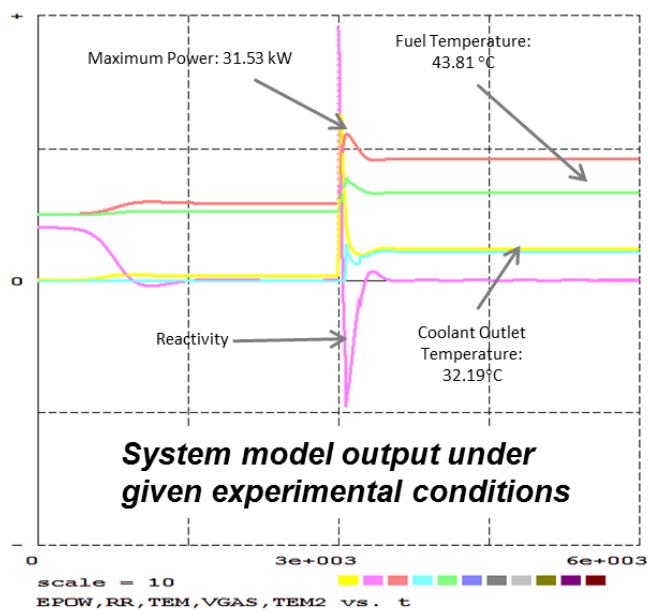


Figure 4: System Model Trace of SUPO Rapid Reactivity Insertion

The trace shows the initial power and reactivity excursion to establish a \$0.10 initial steady-state condition. These conditions were used as system model input. At the 3,000 second point a \$0.48 insertion yields a corresponding rapid increase in both power and reactivity. Fuel temperature and coolant outlet temperature follow the power increase. All subsequently settle to a new steady-state condition. In this example the fission power tracks closely with the reported values while the temperatures scale a bit higher; however, the typical response to a rapid increase in core reactivity is exhibited by SUPO. There is a dynamic region at the time of the insertion followed by the core establishing a new steady-state at a higher power.

Silene: Benchmark for Pulse Operations

Silene was an annular core AHR that employed 93% ^{235}U enriched uranyl nitrate fuel. It operated at the CEA facility at Valduc, France until 2010. Silene was unreflected and possessed no active cooling. Experiments covered the full range of AHR operations grouped as follows:

- Pulse – $\Delta k \gg \beta$; reactivity insertion rate approximately \$20.00/sec
- Slow Kinetics – $\Delta k \leq \beta$; reactivity insertion rate approximately \$0.03/sec
- Free Evolution – reactivity insertion rate approximately \$0.20/sec
- Boiling – $\Delta k \geq \$5.00$; reactivity insertion rate approximately \$0.40/sec

All experimental conditions and results are from Barbry.⁸

Figure 5 includes a photograph and a cut-away drawing of Silene.

⁸ Francis Barbry, CEA IPSN, Report SRSC n° 223-September 1994, Silene Reactor, Results of Selected Typical Experiments, 1994

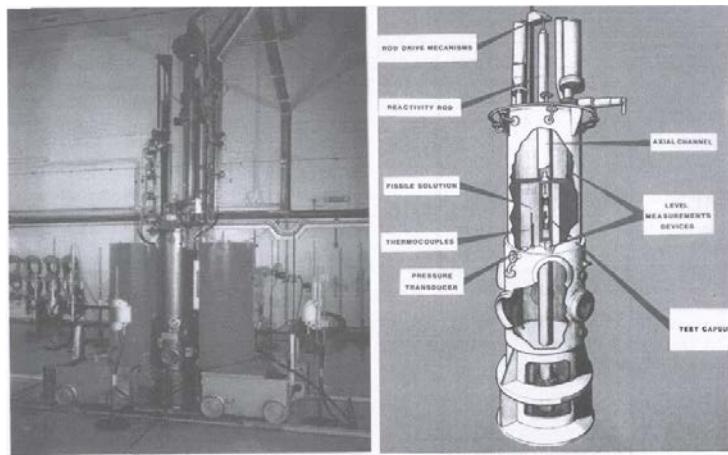


Figure 5: Silene AHR

Silene was particularly well suited for pulse experiments having considerable excess reactivity available and rapid insertion mechanisms. For this reason it is considered the benchmark AHR for pulse type excursions in the super critical region.

Figure 6 is a side-by-side comparison of experiment and system model traces for a \$2.96 pulse of Silene. Both traces are logarithmic in power.

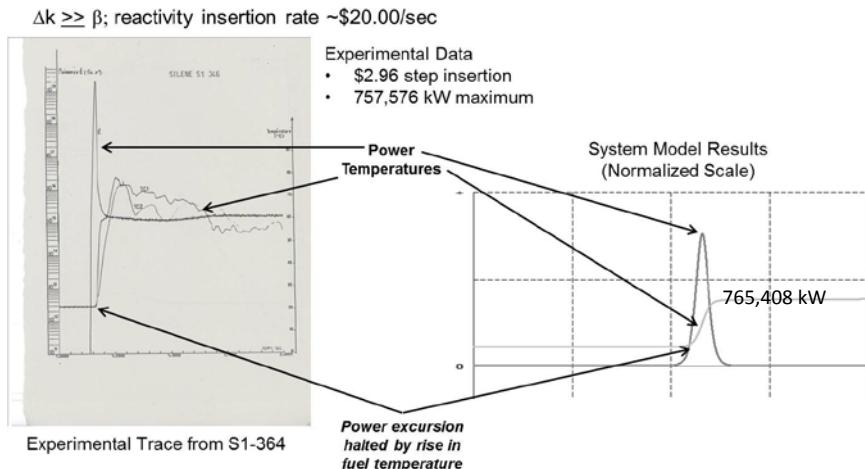


Figure 6: Silene Pulse Operation

Note that the system model result of the given experiment is 765,408 kW peak power compared to the 757,576 kW reported experimental result. The initial peak exhibits the typical shape seen previously in the SUPO results where the power rises until quenched by the onset of temperature rise in the fuel. The experimental trace includes two in-core thermocouples while the system model is reporting a single trace of average fuel temperature; nevertheless the general behavior of the system is the same.

Slow kinetics experiments were performed with total reactivity insertions in the delayed critical region. Figure 7 is an experimental trace from Silene with a \$0.51 reactivity insertion with a side-by-side trace from the Silene system model with the same experimental conditions.

$\Delta k \leq \beta$; reactivity insertion $\sim \$0.03/\text{sec}$

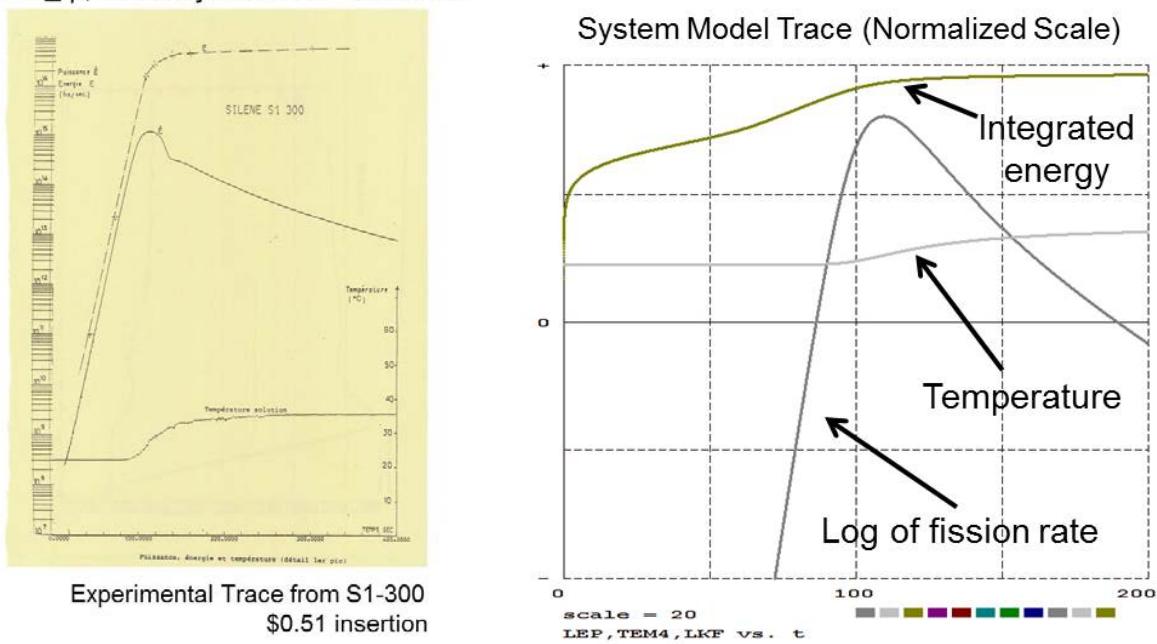


Figure 7: Silene Slow Kinetics Operation

Both traces show the rather gentle response of the system as compared to pulse operations. This response is typical when a fissile solution system experiences a reactivity excursion at a rate that allows the negative temperature feedback response to overcome the thermal inertia of the system on the same timescale. Table 3 compares experimental data to system model results.

Table 3: Silene Slow Kinetics Experimental Data & Model Results

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

Free evolution experiments in Silene are similar to slow kinetics in that the ramp rate is low in comparison to the pulse experiments; however, the total excess reactivity inserted was in the super critical region. Figure 8 is a system model trace of a \$2.96 ramp reactivity insertion of 0.28 \$/sec.

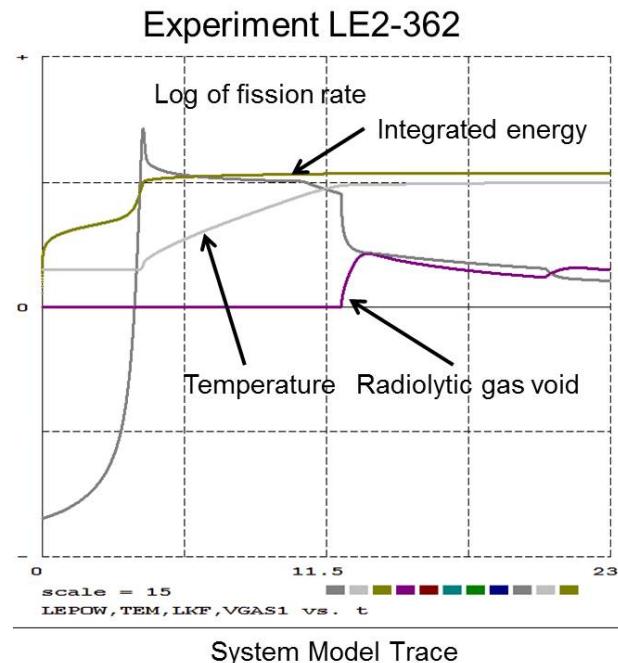


Figure 8: System Model Trace for Silene Free Evolution Experiment

Free evolution experiments contain features of both pulse and slow kinetics operations. Note that pulse experiment S1-364 shown in Figure 6 and the free evolution experiment LE2-262 shown in Figure 8 have the same excess reactivity insertion, \$2.96, but the initial peak in the free evolution case is approximately two orders of magnitude less (see Table 5 below). This is entirely due to the difference in insertion rate: 20.00 \$/sec in the pulse case and 0.28 \$/sec in the free evolution case. In the slow kinetics case, the reactivity insertion rate in the free evolution experiments is sufficiently slow that the core's thermal response can quench the initial peak in a timescale similar to the rise in power, thus halting the rise at a lower value than in the pulse case. Another similarity between the slow kinetics experiment and the free evolution experiment is the broadening of the initial peak. This, again, is due to the slow reactivity insertion rate where the core response is quicker than the rate of insertion; the core responds before all the reactivity has been inserted. This broadens the initial peak. In the pulse case, the opposite is true; the total reactivity has been inserted more rapidly than the core can thermally respond. Taken together the pulse and free evolution experiments show the actual core dynamic response to the reactivity insertion is ultimately driven by the thermal inertia of the core. Additionally, in the pulse experiment the initial peak is quenched long before the core is saturated with radiolytic gas, while in the free evolution experiment the initial peak is finally quenched by the negative reactivity feedback due to the onset of radiolytic gas void. Table 4 compares experimental data with system model results.

Table 4: Silene Free Evolution Experimental Data & System Model Results

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

Boiling experiments are basically free evolution but at a high enough total reactivity insertion to heat the fuel to boiling. Figure 9 presents a Silene system model trace of a \$7.20 reactivity insertion at 0.45 \$/sec.

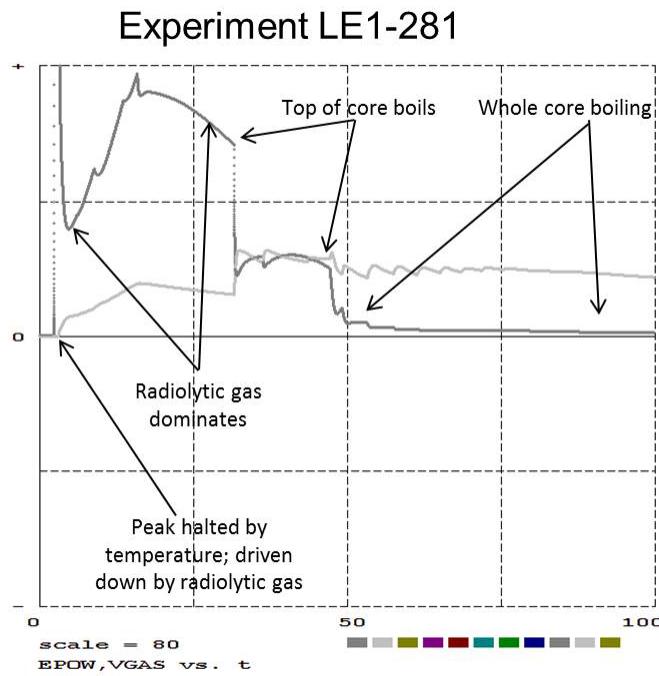


Figure 9: System Model Trace of \$7.20 Ramp Insertion in Silene

The general shape of the curve shown in Figure 9 is similar to that shown for the free evolution experiment shown in Figure 8. There is a large initial peak, but, once again, nearly two orders of magnitude less than the pulse experiment discussed previously even though the total reactivity inserted is more than two times greater in the boiling experiment. The initial peak is halted as before by core temperature rise; however, the amount of reactivity inserted, even at a relatively slow rate, compresses the gas saturation time to occur near the minimum of the initial peak. From this point radiolytic gas void dominates core dynamics until the onset of boiling. The trace in Figure 9 shows a sharp drop in power at the onset of boiling, which occurs near the top of the core. This situation remains nearly constant in power until boiling propagates throughout the core. At this point in time the large void due to steam drives the power down to very low values and the core reaches a steady-state condition characterized by low power, high steam generation rates and the liquid fuel at the boiling point throughout.

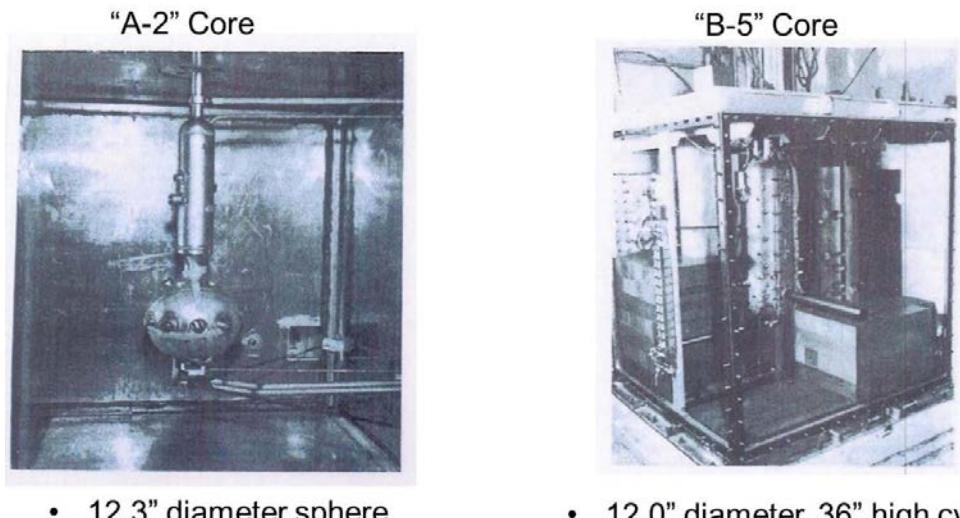
Table 5 compares experimental data and system model results.

Table 5: Silene Boiling Experimental Data & System Model Results

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}

KEWB Steady-State & Pulse Operations

The Kinetics Experiments on Water Boiler (KEWB) were commissioned by the Atomic Energy Commission (AEC) primarily to examine accident scenarios in fissile solution systems. A series of cores were designed and utilized in the KEWB campaign. Figure 10 provides pictures of two. The KEWB "A-2" core was essentially SUPO with higher fuel uranium concentration. Then KEWB "B-5" core was in a cylindrical geometry of the same diameter as the "A-2" core but with essentially twice the uranium concentration in the fuel. The "B-5" core was the test bed for pulse operations and was unreflected, while the "A-2" core, like SUPO, was reflected.



- 12.3" diameter sphere
- 13.7 liters UO_2SO_4
- 106 gU/liter; 93.2% enrichment
- Graphite reflected
- 12.0" diameter, 36" high cylinder
- 13.7 liters UO_2SO_4
- 203 gU/liter; 93.2% enrichment
- Unreflected

Figure 10: KEWB AHR

KEWB "A-2" was operated in both steady-state and pulse modes. Figure 11 shows a trace from a steady-state system model simulation of this core.

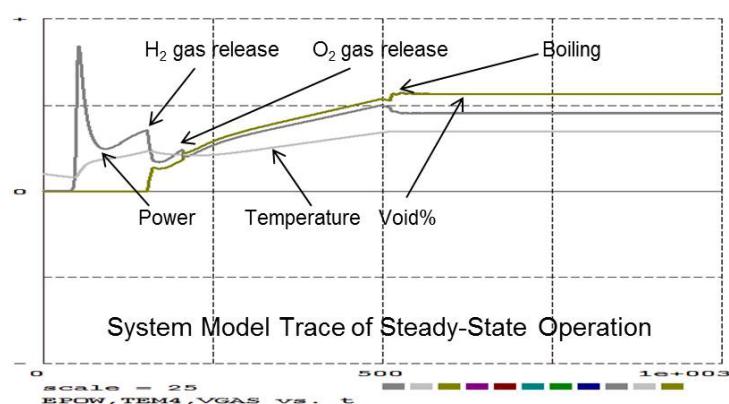


Figure 11: System Model Trace of KEWB "A-2" Steady-State

This trace compares favorably with that for a steady-state operation on SUPO shown in Figure 3. Note also that even though the core does reach a steady-state at boiling the power does not collapse as in the Silene experiment. This is due to the thermal balance in the core being maintained by KEWB “A-2” core cooling, which was not the case in Silene and that the total reactivity inserted in this case is just to cause the onset of boiling while in the Silene core the total inserted amount was more than \$1.00 above the boiling threshold.

KEWB “A-2” was pulsed with reactivity increases that approximated step insertions. Table 6 presents the experimental data on the steady-state operation shown in Figure 11 and a pulse operation compared to system model results.

Table 6: KEWB “A-2” Experimental Data & System Model Results

Operation	Δk_s	Rate (\$/sec)	kW	Temperature
Steady-State	5.00	0.01	50	85
			56.78	87.14
Pulse	3.75	Step	6,500	N/A
			6,470	N/A

KEWB “B-5” was the primary core utilized for the study of pulse behavior. Table 7 presents experimental data and system model results for a variety of step reactivity insertions in the KEWB “B-5” core.

Table 7: KEWB “B-5” Pulse Experimental Data & System Model Results

Δk_s	Experiment Peak Power (MW)	System Model Peak Power (MW)
3.67	4,000	4,072
3.27	2,800	2,941
2.87	2,000	1,680
2.62	1,500	1,149
2.33	1,000	619

Figure 12 presents the system model trace for the \$3.67 step reactivity insertion into KEWB “B-5”.

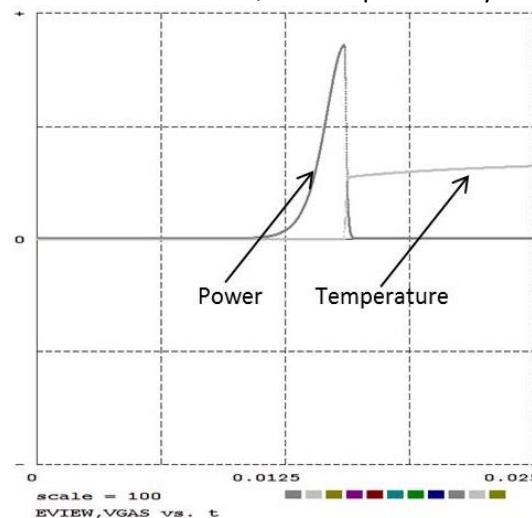


Figure 12: System Model Trace of KEWB “B-5” \$3.67 pulse

Comparing the model trace in Figure 12 with that presented in Figure 6 for a pulse operation in Silene shows the KEWB “B-5” pulse is asymmetric when compared to that of Silene, exhibiting a sharp decrease once the peak is reached. The reason for this is driven by a delayed onset of core temperature rise in KEWB “B-5” as compared to Silene. KEWB “B-5” fuel was at the same ^{235}U enrichment but approximately one-third the volume suggesting the increase in fission energy density occurs much faster in KEWB “B-5” than in Silene due to the uranium concentration. This may be the reason for the delay of the temperature rise in the core as compared to Silene.

HRE – Pressurized Core Benchmark

The Homogeneous Reactor Experiment (HRE) was designed to explore the feasibility of using a solution fueled system to generate electric power. In order to facilitate conversion of the fuel fission energy to steam the desire was to run the core at a high temperature. Since the core was an AHR this required the raising of the boiling point of the fuel. This was accomplished by pressurizing the core and pumping the heated fuel to a heat exchanger.

Modeling pressurized cores required amending the system model by varying material constants of the fuel with temperature, pressure, and salt content, parameters that do not vary significantly in the rather narrow operating range of approximately 40°C ΔT in atmospheric pressure cores. Additions to the generic system model to vary material properties included the following for both fuel and coolant:

- Boiling point
- Thermal conductivity
- Isobaric compressibility
- Expansion coefficient
- Kinematic and dynamic viscosities
- Specific heat
- Thermal diffusivity
- Density

Certain operational parameters required modification as well. These included:

- Radiolytic gas saturation concentration
- Boundary layer thickness
- Radiolytic gas and steam bubble transit times

Comparison of system model results for the amended version 2 to the basic version 1 for unpressurized experiments showed agreement verifying the amendments.

The spherical geometry of the HRE-1 core is shown in Figure 13.

HRE-1 Core



Figure 13: HRE-1 Core

HRE-1 experiments typically produced 1.0 MW at 1000 psi (68 atm). The modified Version 2 of the system model estimates 919 kW at the same pressure. At this operating condition the fuel was just under the 277°C boiling point of water at that pressure.

Summary

Several general conclusions regarding AHR can be made from the forgoing set of experimental and theoretical discussions. AHR can operate in a variety of modes from steady-state to prompt critical depending on the amount of excess reactivity inserted and the rate of that insertion. Peak powers reached in reactivity insertions are generally much higher than ultimate steady-state conditions but, however high, over time a steady-state will be attained. It can be said that any bounded reactivity excursion will result, possibly after a dynamic region, in a new steady-state. This is a definition of stability for fissile solution systems. The overriding characteristics that determine the behavior of fissile solution systems are the inherent large negative reactivity feedback due to fuel temperature and gas void.

These characteristics of AHR offer the engineer a wide design space. Fuel may be essentially any aqueous solution of uranium in concentrations up to the onset of precipitation. Vessel configuration may be tailored to the application since neutronics can accommodate a wide range of geometries. Finally, the operating power of the AHR will be ultimately determined by the ability of the heat removal system to remove the fission generated heat in the core.