Design Principles for Aqueous Homogeneous Reactors (AHR)

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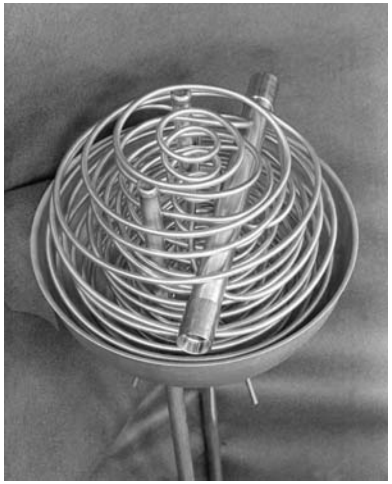
Fissile Solutions and ^{99}Mo Production

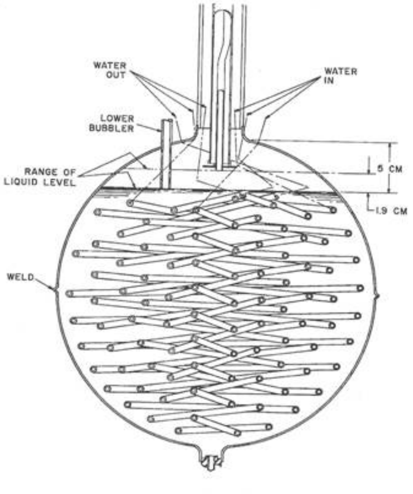
- **Solution Fueled Reactors**
  - In 1944 the third reactor ever built began operation at Los Alamos with Enrico Fermi at the controls; it was a solution fueled reactor called "LOPO" for Low Power
  - Since 1944 – 2005 a series of solution reactors operated nearly continuously at LANL; one SUPO for Super Power operated from 1951 – 1974, is considered prototypical of a Medical Isotope Production Reactor
- **Advantages of Solution Fueled Systems**
  - Inherent stability and safety
  - Relative ease of reactivity control
- **^{99}Mo Production**
  - Approximately 6.3% of fission products from ^{235}U is ^{99}Mo
  - No neutron source required as fissile solution is it's own source
  - No target processing required; ^{99}Mo already in solution

SUPO: Prototypical AHR

Characteristics

- Operated at LANL from 1951-1974
- Spherical, Graphite Reflected, Cadmium & Boron Control Rods, Actively Cooled-30 cm dia.; SS347
- Accumulated ~600,000 kW/h of operation; typically 25kW (1.7 kW/liter) @ 60° C
- HEU Uranyl Nitrate fuel-12.6L; 75gU/L
- Produced ~11 liters/min radiolytic gas @ 25 kW; 2kW/L



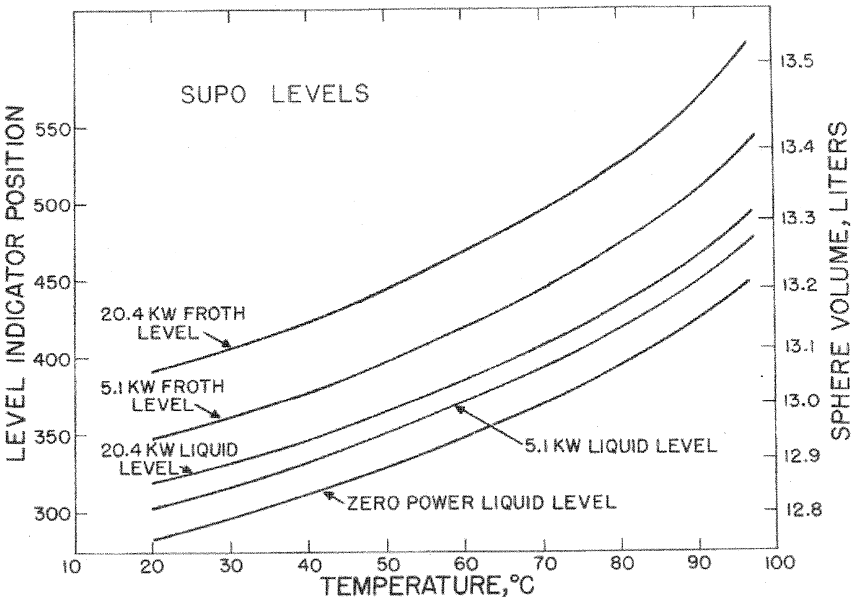


Observations

- Essentially all data on transient behavior of “cold-unsaturated” core; little on steady-state operation of a “hot-saturated” core
- Standard theoretical treatment of transient excursions does 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.

AHR Dynamics

Power Oscillations

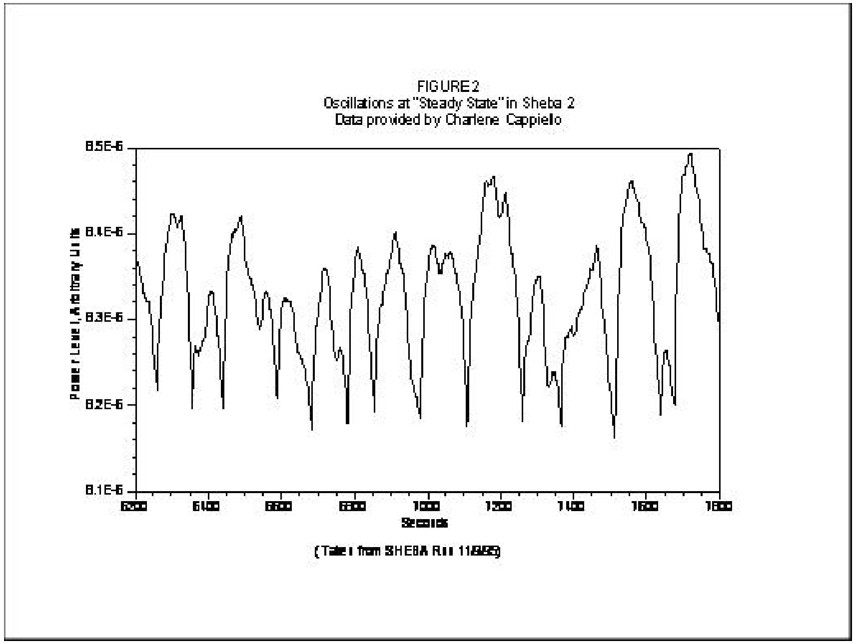


FIGURE 2
Oscillations at "Steady State" in Sheba 2
Data provided by Charlene Cappiello

Variations over Time

- Liquid fuel redistributes itself over very short time scales due to fluid flow
- Fuel density decreases due to fission heating and radiolytic gas void
- Results in strong negative temperature feedback coefficient of reactivity

Theoretical Treatment of AHR at Steady-State

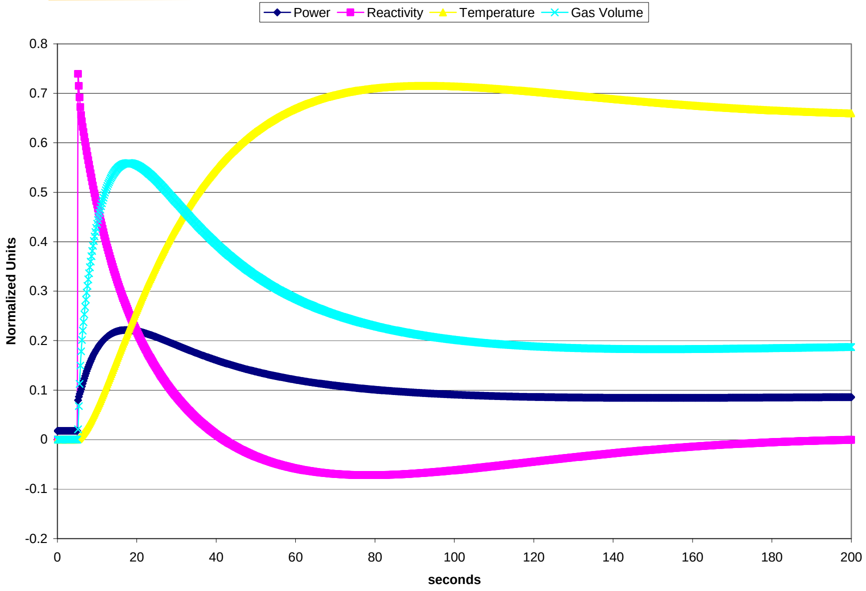
- **SUPO-M a model of SUPO developed to evaluate approaches**
- **Theoretical treatment developed, which closely matches SUPO data when applied to SUPO-M**
- **Model is based on “small bubble” hydrodynamics that are at thermodynamic equilibrium with the solution and rapidly reach terminal velocity and escape the core**
- **Model predicts unconditional stability against step changes in power at steady-state**
- **Major factors important to AHR steady-state performance are related to changes in solution chemistry over time and the effect of those changes on neutronics and gas dynamics**

“Steady-State” AHR Model

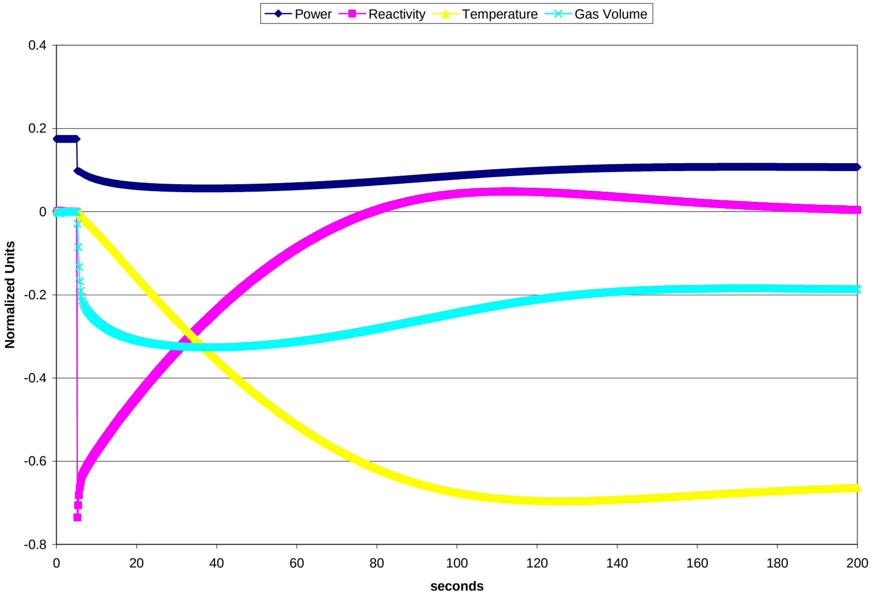
(*Documented in LA-UR-10-04318: “Stability Analysis of the SUPO Reactor”; Kimpland, Hayes, & Grove; June 2010)*

- SUPO empirical data on radiolytic gas production and dynamics could not be reproduced using “standard” theoretical treatments (*Hetrick & Kimpland*), which accurately predict core dynamics of transients from a “cold-unsaturated” core; (*AHR for ^{99}Mo production will operate steady-state with a “hot – saturated” core*)
- Developed new theoretical treatment (spatially independent), which closely matches SUPO performance
- Model is stable (stability refers to the characteristic that an AHR initially at steady state re-establishes a new steady state condition on its own following a reactivity perturbation); model response to changes very docile

Stability Predicted by New Model



Model response following a $0.75 step in the SUPO reactor with an initial steady state power of 25 kW.



Results for a $-0.75 step insertion from an initial power of 25 kW.

Model predicts for well designed cores any available step in reactor power will result in a new steady-state (no unbounded excursions)

Fuel Chemistry Effects of AHR Reactivity

(Documented in LA-UR-10-04317: “Effects of Solution Chemistry on Aqueous Homogeneous Reactor (AHR) Klein; June 2010)

- General mechanisms include
  - Dynamics – Indirect effect due to gas transport and heat removal (viscosity, specific heat)
  - Neutronics – Direct effect due to elemental composition over time (Water loss, Nitric acid radiolysis and nitrogen depletion, pH variation, Burn-up)
- **Conclusions**
  - Solution chemistry strongly affects reactivity (Hydrogen, Nitrogen & Fission Products)
  - Solution chemistry including water make-up, pH stability, and related factors must be part of any AHR control system

AHR Design Considerations (*State of the Art*)

- **Lack of empirical data on steady-state (hot, saturated) behavior forces design dependence on general theoretical principles and anecdotal inferences from historical record of transient conditions**
  - Based on theoretical computations of reactivity, considerable excess reactivity must be present to handle reactivity variations over time from radiolytic gas production and fuel chemistry changes due to pH, water content, fission product inventories
  - Off-gas handling system must be capable of handling approximately 0.44 liters/min/kw of Hydrogen + Oxygen in an approximate stochiometric mixture and approximately 2.5 cc/min/kw of mixed oxides of nitrogen (*King, 1955*)
- **If cooling is provided internal to the core design of cooling tubes or coils should not impede the transport of radiolytic gas from the system (*Kimpland & Hayes, 2010*)**

Concluding Remarks on AHR Development

- While technology had largely been demonstrated over decades the peculiarities of operating at steady-state (hot, gas saturated core) has not been assessed either theoretically or empirically
- Effects of full fuel cycle are only now being examined; limits on uranium concentration, core chemistry, presence of fission product inventories as related to core reactivity and separations efficiency largely unknown
- Greatest impediment to realizing an operational AHR for isotope production is the lack of empirical data