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Design Principals for Aqueous Homogeneous Reactors

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ABSTRACT

Los Alamos National Laboratory developed and operated reactors using fissile solution fuels for over 65 years. The primary purpose of these reactors was to provide a reliable and predictable neutron flux for experimental nuclear physics, detector, and alarm evaluation; however, surprisingly little detail exists on reactor operating characteristics, particularly at steady-state. What data is available has been catalogued and extended by theoretical treatment to develop a prescription for successful design for aqueous homogeneous reactors (AHR) to produce ^{99}Mo . Approaches to management of radiolytic gas, temperature effects, and impact on stability are discussed. In addition results of recent experiments on a variety of fuel types are presented.

INTRODUCTION

At the dawn of the nuclear age over six decades ago little experimental data was available to test the theoretical concepts emerging in nuclear physics. At Los Alamos National Laboratory it was realized that a reactor fueled with uranium in solution could be rapidly produced and would likely provide a sufficient neutron flux to address the major questions related to cross section and critical mass. Hence, the third reactor ever built was a uranyl sulfate fueled reactor, dubbed LOPO, for Low Power. LOPO was placed into operation at Los Alamos in 1944 with Enrico Fermi at the controls.

Soon thereafter a second reactor, HYPO (High Power), was constructed, followed by a third, SUPO (Super Power). Both were fueled with uranyl nitrate, since it was realized that uranium metal was easier to dissolve in nitric acid. SUPO operated at LANL from 1951 until 1974, amassing over 600,000 kWhr of operation. A picture and cross section diagram of SUPO is shown in Figure 1.

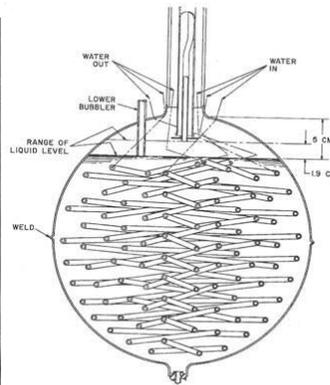
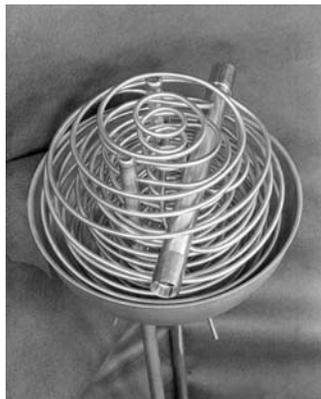


Figure 1: SUPO

The basic features of any solution fueled reactor can easily be seen. A reaction vessel holds the fuel; in the case of SUPO a 12" diameter sphere. Immersed in the fuel are cooling tubes. SUPO used coils for this purpose. Above the fuel is a void, basically serving the dual purpose of expansion chamber for the fuel as it heats up and a plenum for the large amounts of radiolytic gas formed during operation.

What is remarkable about the historical record on the operation of SUPO and other solution fueled reactors is that little empirical data exists on the characteristics of the reactors themselves, while considerable detail exists on the neutron flux produced. An example is shown in Figure 2.

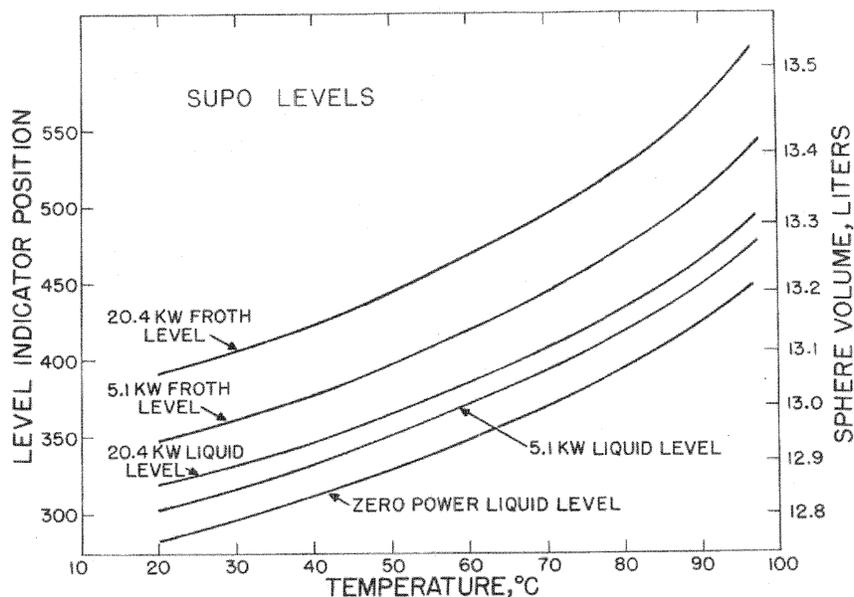


Figure 2: SUPO Operational Data

This graph, reproduced from King¹ is the single piece of data found in the historical record showing volume change, and the existence of a foamy layer at the top of the core, as a function of operating temperature. At first glance this graph provides insight into the void fraction caused by radiolytic gas bubbles but how many data points were used to construct the curves? And, what are the errors in each entry? Neither the publication nor the logbook contains this information leading one to conclude that these curves are simply analytic functions.

King reports other data that is tantalizing in its brevity. Three examples being:

- “After the HYPO had been run for several hundred kilowatt hours, it was observed that its reactivity had increased remarkably.”

¹: L.D.P.King, International Conference on the Peaceful Uses of Atomic Energy, “Design and Description of Water Boiler Reactors”

- “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.”

Examination of the logbooks indicate that SUPO typically operated a few hundred to at most a few thousand seconds, a rather short time compared to the 5 days minimum continuous operation required of an Aqueous Homogeneous Reactor (AHR) in the current vernacular.

A video exists showing the dynamic behavior of SUPO even at rather low power densities. This grainy video shows considerable bubbling on the surface of the solution across the entire operating temperature range of 40°C - 80°C. No doubt this vigorous foaming and bubbling was the genesis of the term “Water Boiler” used to describe these reactors over the years, even though no boiling ever occurred in any of these systems.

The dynamic nature of solution reactors is illustrated in a power graph, shown in Figure 3, taken at LANL in 1995 during a rather long run (~7000 seconds) of Sheba.

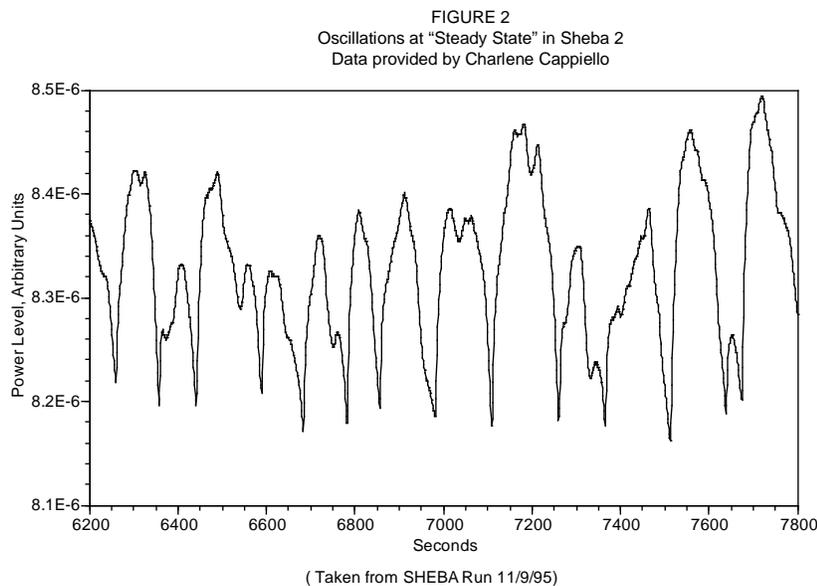


Figure 3: Sheba “Steady-State” Power

This graph is typical solution reactor operation at “steady-state”, which may be defined as a “hot” core saturated with radiolytic gas.

The principal characteristic of a solution fueled reactor that distinguishes it from all others employing solid fuel is that the fuel is dynamic over extremely short time scales due to the ability of a liquid to reconfigure itself rapidly by fluid flow. It is precisely this characteristic that results in a strong negative temperature feedback coefficient of reactivity, drives inherent stability, but requires considerable care in operation.

THEORETICAL CONSIDERATIONS

Kimpland, Hayes, and Grove² undertook a theoretical analysis of SUPO in an attempt to understand the characteristics of radiolytic gas formation and transport by developing a gas bubble hydrodynamic model for the reactor operating at steady-state. Average bubble velocity as a function of height in the core and the void fraction as a function of time, respectively, resulted from that analysis as well as estimates of average void fraction for any assumed bubble size.

SUPO operational data provided in Figure 2 at 20 kW suggests a void fraction of approximately 0.9% due to radiolytic gas at that temperature. The new gas bubble hydrodynamic model suggests that an average bubble size of approximately 0.25 mm reproduces this empirical data. The model also predicts that the void fraction in the core increases linearly with core height and that the dimensions of radiolytic gas bubbles are considerably smaller than those of steam bubbles and that once formed quickly reach local thermodynamic equilibrium with the surrounding solution. This theoretical treatment was extended to examine stability of an AHR. In this context stability refers to the characteristic that an AHR, initially at steady state, re-establishes a new steady state condition on its own following reactivity perturbation.

A nonlinear reactor system model for a “water-boiler” was proposed by Skinner and Hetrick in 1957³. This model uses space-independent kinetics equations that include the effects of delayed neutrons, temperature, and radiolytic gas. Kimpland et al, the Skinner/Hetrick model has been modified to incorporate space-dependent gas transport and the use of the normalized point reactor kinetics equations.

The simulation model is used to perform a “set point” analysis, where the reactor system is initially at steady state and a reactivity perturbation, in the form of a step insertion, is introduced. The simulation model then tracks the change in system state-variables from initial conditions. The simulation model has been used to investigate a wide range of transients at various initial steady state power levels and with various reactivity perturbations. For the purpose of illustrating the basic behavior of the model, a series of transients initiated with a reactivity step insertion of \$0.75 is presented. Figure 4 shows the model’s prediction of power, following a \$0.75 step insertion at 5 seconds, for the SUPO reactor initially at a steady state power of 25 kW.

² LA-UR-10-04318: “Stability Analysis of the SUPO Reactor”; Kimpland, Hayes, & Grove; June 2010

³ R Skinner and D. Hetrick, “Transfer Function of a Water Boiler Reactor,” Atomic International Document NAA-SR-1948, August (1957).

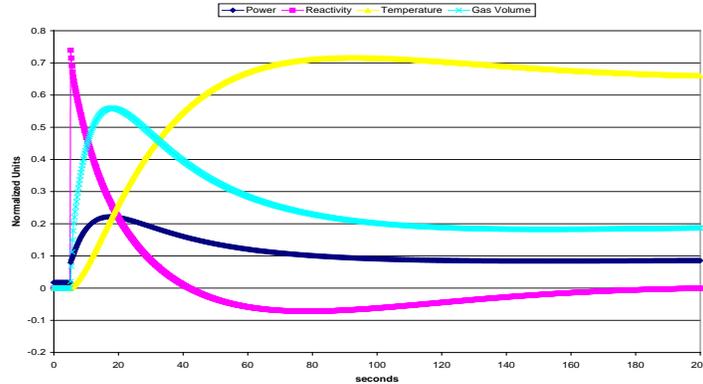


Figure 4: Stability of SUPO Model with \$0.75 Step Reactivity Increase

This result shows the prompt step in reactivity is followed by a return to a steady-state condition. Temperature, power, and gas volume track this pattern. It may be concluded that for well designed cores any available step in reactor power will result in a new steady-state (no unbounded excursions).

FUEL CHEMISTRY

Klein⁴ summarized the various effects of fuel chemistry on AHR solution reactivity concluding that in both the dynamics of the core and neutronic effects are strongly dependent on fuel chemistry. Indirect effects are due to gas transport and heat removal (viscosity, specific heat). Neutronics is directly affected due to elemental composition over time (Water loss, Nitric acid radiolysis and nitrogen depletion, pH variation, Burn-up).

In 2011 May⁵ performed separations after irradiation of uranyl nitrate and sulfate samples of up to 300 gU/l with approximately 90% recovery. This result suggests that uranyl sulfate is a viable fuel for AHR for ⁹⁹Mo production.

CONCLUSIONS

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, and fission product inventories.

Off-gas handling system must be capable of handling approximately 0.44 liters/min/kw of Hydrogen + Oxygen in an approximate stoichiometric mixture and approximately 2.5 cc/min/kw of mixed oxides of nitrogen (in uranyl nitrate systems)

Uranyl sulfate fuel can be used for AHR applications for ⁹⁹Mo production.

⁴ LA-UR-10-04317: "Effects of Solution Chemistry on Aqueous Homogeneous Reactor (AHR) Klein; June 2010

⁵ I. May; unpublished results

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.

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