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Management of Criticality in Subcritical Solution Systems

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ABSTRACT

Systems have been proposed to produce ^{99}Mo employing an accelerator to induce fissions in a subcritical configuration of fissile solutions. Historically, k_{eff} has been used to judge the "stand-off" from critical with 0.95 being chosen as the point of reference. However, it is shown that for a variety of configurations k_{eff} is not a useful estimate of the physical margin from critical; rather a directly measureable quantity such as solution volume provides a reliable means for this purpose. Examples of this principle are discussed along with operational control mechanisms.

INTRODUCTION

Fissile solution systems of uranium have long been used as a reliable, and predictable, source of neutrons for experimental nuclear physics, dosimetry, and criticality alarm calibration. Los Alamos National Laboratory (LANL) operated solution reactors employing fissile uranium fuel for nearly 65 years; indeed, the third reactor ever built was a solution fueled reactor. This reactor, dubbed "LOPO" for Lo̲w Po̲wer, started operations at LANL in 1944 with Enrico Fermi at the controls. The third generation of this system, SUPO, for Su̲per Po̲wer operated from 1951 until 1974 and is considered by many as prototypical of a solution reactor of this class. The picture and cross section diagram of SUPO presented in Figure 1 shows typical features of a solution reactor, or Aqueous Homogeneous Reactor (AHR) in the current vernacular.
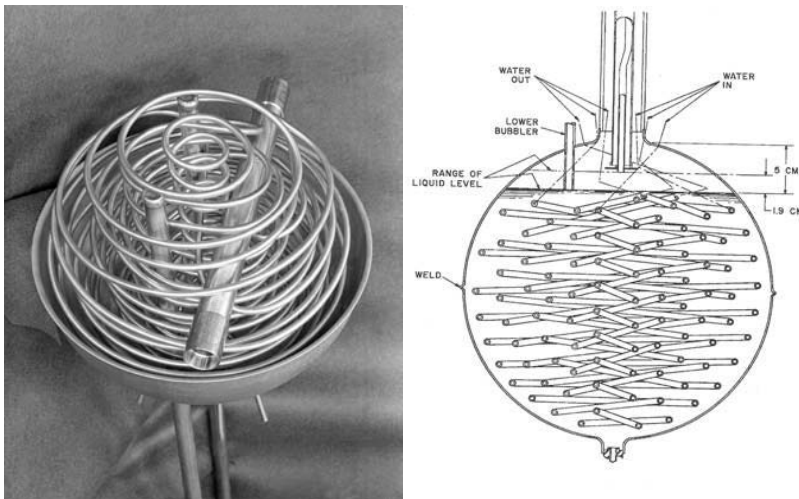

Figure 1: SUPO

A reaction vessel, in this case a spherical bowl, holds the fissile solution. Immersed in the fuel are cooling coils to maintain the solution temperature below boiling. A head space, provided above the fuel, serves as both an expansion chamber and a plenum for the radiolytic gas produced during operation. With the exception of those developed only for transient operation where cooling was not required, essentially all solution fueled reactors incorporate some variation on this configuration. Certainly, any modern AHR developed for production of [99]Mo, which requires nominally five days of steady-state operation, must be cooled continuously.

REACTIVITY EFFECTS IN SOLUTION SYSTEMS

The single characteristic that distinguishes fissile solution fueled systems from all other reactors is the dynamic reconfiguration of the fuel during operation. Being a liquid, the fuel changes physical parameters such as occupied volume and density and continuously redistributes through fluid flow.

Temperature effects driven by fission power in the solution increases fuel volume and decreases density resulting in lower reactivity. This is the historically recognized negative temperature coefficient of reactivity of fissile solutions. The reactivity feedback due to thermal effects may be quite significant; however, thermal inertia of solutions results in a rather long time constant related to this reactivity change. Feedback due to density and volume fluctuations associated with radiolytic gas formation in the solution may, however, be quite dynamic.

Early observers, witnessing the dynamic behavior of solution reactors, called them "water boilers" even though their operating temperature remained far below the boiling point. SUPO, operating at modest power densities of less than a kilowatt per liter, was seen to bubble vigorously even at solution temperatures of only 40°C. Radiolytic gas generation results in a stratification of fuel density as the gas bubbles rise under buoyancy and the appearance of a foamy layer on the solution surface. Even at modest power densities radiolytic gas causes rapid reactivity oscillation of the system.

The vast majority of gas produced in a fissile solution system is hydrogen and oxygen produced by water radiolysis by fission products. King[1] reported the amount of this radiolytic gas to be approximately 0.44 liters/minute/kW. An AHR of operating at 200 kW, typical for those considered for [99]Mo production, will have to manage 88 liters of radiolytic gas per minute or 633,600 liters in a five day [99]Mo production cycle. Thus, continuous water makeup is required. SUPO operating at 25 kW lost approximately 0.5% of its reactivity in only two minutes in the absence of water makeup.

Changes in solution chemistry over time also contribute significantly to reactivity feedback in fissile solution systems. SUPO employed uranyl nitrate fuel as is being considered for [99]Mo production solution fueled systems. King[1] again reports a loss of approximately 2.5 cc/minute/kW of nitrogen in SUPO. Addition of 185 ml of 15.8 Normal HNO_3 was required to replace loss (equal to 10 g [235]U or $0.37) every 1000 kW hours of operation. Reactivity control in uranyl nitrate fueled systems thus requires acid makeup along with water addition. This is one reason for the interest in uranyl sulfate as a fuel, which does not exhibit reactivity feedback due to acid loss.

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[1] Design and Description of Water Boiler Reactors, L. D. P. King, International Conference on the Peaceful Uses of Atomic Energy, 20 June 1955

Over a longer term, solution fueled systems suffer a reactivity loss due to uranium burn-up and build-up of fission product inventories. In a single year this loss can amount to 20% or more of total reactivity in the system.

The conclusion that can be drawn from these brief observations is that reactivity effects in fissile solution fueled systems are quite dramatic, amounting to measureable percentages of total system reactivity, with time constants ranging from seconds to months, all of which must be managed to optimize the performance of a production system.

REACTIVITY IN SUBCRITICAL SOLUTION FUELED SYSTEMS

In an AHR long term changes in reactivity due to ^{235}U depletion or the presence of fission product inventory can be largely managed by providing and controlling excess reactivity above delayed critical. Control rods made of neutron absorbing materials such as boron or cadmium have historically been used for this purpose as they can be withdrawn to "add" reactivity as needed to maintain the desired operational value.

In AHR the excess reactivity over delayed critical is chosen to manage system operational parameters such as power fluctuation and stability. Subcritical systems, however, must manage reactivity to balance the desire to maximize production, as measured by fissions per source neutron in accelerator-driven systems, while maintaining stand-off from delayed critical. A typical value chosen by the accelerator and regulatory community for this stand-off has been an effective reactivity, k_{eff}, ≤ 0.95. However, as discussed below this value may not be a reliable measure of the physical margin from delayed critical for a wide class of subcritical systems.

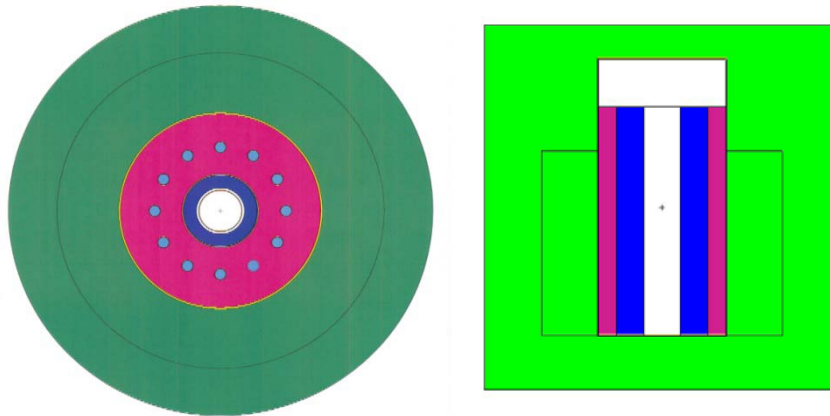Consider the system configuration shown in Figure 2.

Figure 2: Subcritical Solution Fueled System Configuration

The centermost region in this configuration would house structures associated with the accelerator and other than their physical size and composition contribute little to the reactivity of the system. External to this region is a solution fuel annulus with embedded cooling pipes. The exterior annular region serves as both moderator and reflector.

The design evolution of this configuration explicitly establishes the stand-off from delayed critical. This process includes the following steps.

- Selection of a candidate Low Enriched Uranium (LEU) fuel
  - Uranyl nitrate or sulfate
  - Uranium concentration in grams per liter
- Select a candidate vessel configuration
  - Height
  - Annular dimensions
  - Placement, material, and static void due to cooling pipes
  - Coolant, reflector, moderator material
- Estimate the volume at delayed critical for a "cold" (20°C) fuel
- Reduce fuel volume by 5%
- "Turn on" accelerator and estimate fissions per source neutron for the "hot" (60°C) fuel

Estimates of fissions per source neutron are made by adjusting solution density and height in the reaction vessel and applying appropriate changes to model parameters.

A specific example of the above configured system utilizes 90 gU/liter uranyl sulfate fuel in a vessel of 100 cm height and approximately 30 cm in annular radius (i.d. 27 cm; o.d. 57 cm). The coolant and reflector/moderator is heavy water. In this configuration the solution volume at delayed critical is approximately 451 liters; hence, according to the above prescription the 5% volume reduction would be approximately 23 liters of fuel to 428 liters. Note that this is a substantial physical stand-off from delayed critical that can easily be managed by either limiting the volume of available feedstock to this value or by vernier control of fuel height.

The question of course is to estimate the k_{eff} of this system. Due to the design approach, $k_{eff} = 1.0000$ at 20°C for the full 451 liters of fuel. What is remarkable is that removal of 23 liters of fuel depresses k_{eff} only to 0.9974, which appears to be quite near delayed critical, regardless of the rather large physical stand-off of 23 liters. If one asks what volume of fuel represents the typical 0.95 value for k_{eff} it is found to be approximately 250 liters of total fuel; only 55% of the 451 liters at delayed critical. Figure 3 presents k_{eff} for this configuration at various volumes.
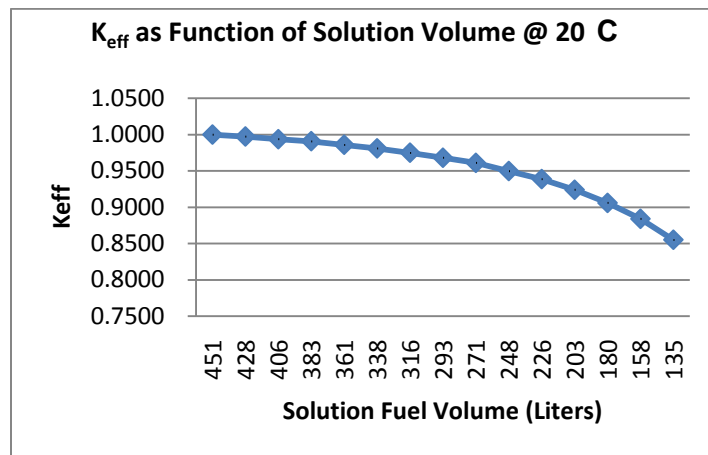

Figure 3: Reactivity as Function of Solution Volume in Annular Core System

All of the above reactivity values are computed for a "cold" 20°C core. Increase in solution volume and decrease in density at operation will depress these values. It is instructive to note that at the full 451 liters fuel volume the reactivity at 60°C is 0.993, the equivalent of approximately 50 liters of fuel reduction over the critical volume.

Over the past year LANL has examined a wide variety of annular core systems of this nature with both uranyl nitrate and sulfate fuels and this is a typical result; large physical reductions in fuel volume result in very minor changes in k_{eff} of the system.

The fundamental reason for this characteristic is that the basic design of these systems purposefully departs from a configuration that is ideal for an AHR where the height to diameter ratio is generally kept close to one. These systems have the geometry where the annulus thickness is considerably smaller than the height of the vessel and the reactivity worth of the fuel is kept low by employing low uranium concentration fuels.

STARTUP AND OPERATION OF SUBCRITICAL SYSTEMS

Startup and operation of this class of assemblies takes advantage of the basic characteristics of the liquid fuel system where volume can be controlled with high fidelity; fuel height in solution reactors operated at LANL was routinely controlled to one millimeter. In the present example this corresponds to less than ½ liter of fuel.

The traditional startup process for solution systems is to perform a "1/M Approach to Critical" (where "M" is multiplication). Figure 4 is a 1/M plot for the example system.
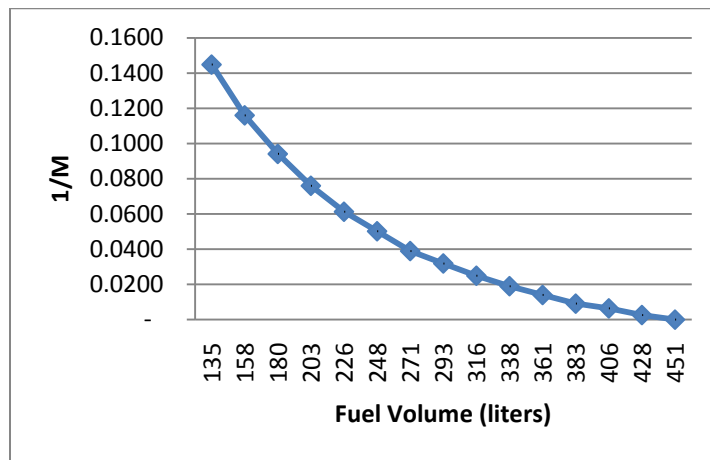

Figure 4: "1/M" Plot by Fuel Volume

Note the system remains conservative throughout the volume range; any estimate for critical fuel volume using any two consecutive estimates is lower than the actual volume at critical, hence, employing the "half-way rule" where an operator is limited to volume addition of ½ the estimated volume change to critical would still result in the system remaining physically far subcritical. In an actual startup process filling the system to 90% of the estimated critical volume as an initial starting point is 406 liters, which is 45 liters from critical, even though the k_{eff} of the system at this point is approximately 0.994. A volume increment of even 10 liters at this point would provide an accurate estimate of critical while remaining physically well below critical. Indeed, the physical margin from critical at 95% of the estimated critical volume is sufficient to allow an approach to

critical by addition of 1 liter of fuel per increment, and finally a few millimeters if the desire is to exactly reach delayed critical.

The key point in this thought exercise is that this approach to critical is conducted with a "cold" core, thus representing the maximum reactivity that the system could ever achieve. Once delayed critical is reached turning on the accelerator immediately results in a substantial margin from critical. In the example given, as noted above, fuel heating resulting from this alone is the equivalent of loss of 50 liters of fuel below critical.

The conclusion that is drawn is that this class of system can be easily and safely operated at very high k_{eff} since the physical standoff from critical is very large. Furthermore, the nature of these systems is such that when driven by the accelerator they depart dramatically from critical. Clearly, for these systems it is reasonable to argue that k_{eff} is not a useful measure of the physical margin from critical.