The Development of a Small Inherently Safe Homogeneous Reactor for the Production of Medical Isotopes

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Summary

The use of radioisotopes for various procedures in the health care industry has become one of the most important practices in medicine. New interest has been found in the use of liquid fueled nuclear reactors to produce these isotopes due to the ease of fuel processing and ability to efficiently use LEU as the fuel source. A version of this reactor is being developed at the Royal Military College of Canada to act as a successor to the SLOWPOKE-2 platform. The thermal hydraulic and transient characteristics of a 20 kWt versionare being studied to verify inherent safety abilities.

1. Introduction

The development of small, inherently safe, nuclear fission reactors for radioisotope production could help to alleviate a supply shortage of radioisotopes once the NRU shuts down. Renewed interest in the aqueous homogeneous reactor has been found for accomplishing this task due to its liquid fuel nature. Liquid fuel solves a number of the fuel and isotope separation problems that have kept isotope production costs high, and also helps with the current need for the use of HEU. Of particular interest here is the development of a small, inherently safe aqueous homogeneous reactor that's primary task would be isotope production but that could also provide research benefits to a university or laboratory. A number of these reactors actively producing radioisotopes would help provide a dispersed source that could serve locations closer to the reactor. A 20 kWt homogeneous reactor is in development at the Royal Military College of Canada as a replacement for the SLOWPOKE-2 reactor.

2. The Aqueous Homogeneous Reactor Concept

The development of the homogeneous reactor which combines moderator/coolant and uranium fuel in liquid solution was among the first nuclear systems to be developed after the discovery of fission. Renewed interest in the technology has been spurred in an effort to produce radioisotopes cost effectively and without the use of HEU. An aqueous homogeneous reactor is very different in design from a traditional heterogeneous reactor with solid uranium oxide fuel.

The homogeneous reactor relies on the use of a liquid fuel consisting of a solution of enriched uranium salt, traditionally either uranyl nitrate $(UO_2NO_3)_2$ or uranyl sulfate (UO_2SO_4) , acid to repress hydrolysis in the solution, and water (moderator/coolant). The flexibility of the reactor design is a very important attribute that aqueous homogeneous reactors (AHR) possess. This flexibility makes it possible for a heterogeneous core to be replaced with a homogeneous core with minimal modifications to original core dimensions.

The reduction in the amount of uranium needed on site and the increase in molybdenum extraction efficiency has also contributed largely to the attention that is being given to AHR's. An AHR uses its fission fuel to produce radioisotopes rather than a fabricated target. This provides a huge savings in uranium requirements and target fabrication costs as well as reduces the amount of uranium required per curie of ⁹⁹Mo[1].

3. Reactor Design

The homogeneous SLOWPOKE design described here consists of fuel solution containing uranyl sulfate (UO_2 -SO₄) and light water (H_2O) contained in a single cylindrical zircaloy containersurrounded by a beryllium reflector and is designed for a nominal 20 kW of heat production. The reactors design is based on the dimensions of the SLOWPOKE-2 in an effort to cut initial cost of installation of the reactor when replacing the SLOWPOKE-2 (Figure 1). The radius of the reactor was therefore fixed to be able to fit within the reflector cavity of a SLOWPOKE-2.



Figure 1 – Left: Section view of homogeneous SLOWPOKE reactor drawing. Right: SLOWPOKE-2 reactor drawing

Initial neutronic calculations showed that reactor criticality could not be met without transgressing the solubility limit of uranyl sulfate in water so the axial height of the reactor hasto be increased to roughly double that of the SLOWPOKE-2in order to obtain a critical mass [2].

4. Transient Analysis

To fully test the inherent safety of the proposed homogeneous reactor, transient simulations are needed to insure that the reactor will not enter an unsafe state in all situations. The use of COMSOL Multiphysics, a finite element analysis tool, was employed here to simulate heat transfer, fluid momentum, and neutron point kinetics.

4.1 Thermal Hydraulic System

The first step in the modelling process was to design the thermal hydraulic system to accommodate the fission heat generation. Like the SLOWPOKE-2, the homogeneous SLOWPOKE will use natural convection of the reactor pool water to cool the core. This system is simulated in COMSOL using coupled mass transport and fluid dynamics physics to model the thermal hydraulic properties. The momentum transfer, continuity equation, and heat transfer equations (as applied in COMSOL[3])used are shown below, respectively.

$$\rho \frac{\partial u}{\partial t} + \rho u \cdot \nabla u = -\nabla p + \nabla \cdot \mu \Big((\nabla u + (\nabla u)^T) \Big) - \rho g$$
$$\frac{d\rho}{dt} + \nabla \cdot (\rho u) = 0 \quad , \qquad \rho C_p \Big[\frac{\partial T}{\partial t} + u \cdot \nabla T \Big] = \nabla (k \nabla T) + Q_f$$

where: p = Pressure, μ = Dynamic Viscosity, g = Acceleration due to gravity, P= Density, C_p = Heat Capacity, T = Temperature, u = Velocity Field, t = Time, k = Thermal Conductivity, Q_f = Heat Source, from fission and decay.

The coolant inlet orifice size was adjusted to find an optimal size to allow for sufficient cooling without altering the neutronic characteristics of the reactor. The exit channel geometry was also altered from having a 90° bend and exiting horizontally, to having a straight "chimney" type exit out the top of the upper reflector. The thermal output from COMSOL is shown in Figure 2 as well as the comparison of the cooling ability of various inlet channel sizes for both the horizontal cooling channel outlet and the "chimney" cooling channel outlet.

4.2 Point Neutron Kinetic Analysis

The first priority of these simulations is to confirm the inherent safety of the homogeneous SLOWPOKE reactor in all reasonably possibly scenarios. This is accomplished by using point neutron kinetics to calculate the nuclear parameters of the reactor during transient situations. Reactivity coefficients calculated by Gagnon[4] using WIMS-AECL[5] and MCNP[6]have been used within the point kinetic (PK) equations to allow the model to respond to reactor temperature and fuel solution state.

The power and temperature of the reactor were key properties which were monitored to determine if both stay within the safety margins of the materials used to contain the radioactive

fuel. The point kinetic neutron density and delayed neutron precursor concentration equations used are shown below [7], respectively.

$$\frac{dn}{dt} = \frac{(\rho - \beta)}{\Lambda} n + \sum_{i=1}^{6} \lambda_i C_{n,i} \quad , \quad \frac{dC_{n,i}}{dt} = \frac{\beta_i}{\Lambda} n - \lambda_i C_{n,i}$$

where: β_i = fraction delayed neutrons in group i, $C_{n,i}$ = delayed neutron concentration in group i, Λ = average neutron generation time, l = neutron lifetime.



Figure 2 – Left: Axial symmetric COMSOL thermal image of the SLOWPOKE "chimney" cooling system. Right: Average reactor temperature for various inlet orifice heights for both cooling channel exit geometry at 20 kWt.

To determine if the reactor stays within safe parameters during transients started from shutdown conditions (system temperature 20°C, sub-critical), various excess reactivities were inserted and the reactor characteristics recorded (Figure 3). Further tests are still in progress.

4.3 COMSOL Neutron Diffusion Analysis

To further the accuracy of the reactor simulations, the use of neutron diffusion codes have been implemented in a survey simulation. Diffusion physics introduces spatial variations in flux and can potentially provide a more accurate neutronic simulation of the core to be coupled with heat and momentum equations. The general diffusion equation [8] applied and a plot of the preliminary results for thermal flux and fast flux of the reactor are shown below.

$$\frac{\partial n(r,t)}{\partial t} = D\nabla^2 \phi(r,t) - \Sigma_a \phi(r,t) + S(r,t)$$

where: n = neutron density, r = vector point in system, t = time, D = neutron diffusion coefficient, \emptyset = neutron flux, Σ_a = macroscopic cross section for absorption, S = Neutron source.



Figure 3 – Left: Power transient results for various reactivity insertions to the homogeneous SLOWPOKE @ 20°C. Right: Thermal and fast normalized neutron flux diagram of axial symmetric SLOWPOKE reactor.

5. Conclusion

From the initial transient results using the PK neutron equations, it can be seen that the SLOWPOKE reactor responds within safe parameters to every possible reactivity insertion to the system. Only with the maximum insertion of 5.872 mk does any part of the fuel solution approach boiling temperatures of 100°C. Further tests will be conducted to test for incidences such as loss of coolant accidents and control rod scram scenarios.

The potential of COMSOL to use neutron diffusion equations to model a nuclear reactor core seem promising from the survey simulations taken thus far. Further implementation of the diffusion equations coupled with heat and momentum equations will be made to see if a time dependent model can be simulated for more accurate transient analysis.

6. References

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[3] COMSOL Multiphysics ®, "COMSOL documentation", Version 4.3a.

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[6] Monte Carlo Group, "MCNP5 - Monte carlo N-particle transport code version 5", Los Alamos Laboratory, Los Alamos, New Mexico.

[7] Glasstone, Sesonske (1967), "Nuclear reactor engineering", 3^{rd} Edition, Equations 5.9 – 5.10, pages 231 - 232.

[8]Ibid.,Equation 3.7, pg. 133.

^[5]Atomic Energy of Canada Limited, "WIMS-AECL 3.1", Reactor and Radiation Physics Branch, Chalk River Laboratory, Chalk River, Ontario.