Safety Analysis of a Homogeneous SLOWPOKE Reactor

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ABSTRACT

A homogeneous SLOWPOKE reactor concept for the production of medical isotopes (such as Molybdenum-99) is being designed at the Royal Military College of Canada. The reactor core was designed as a tank with the diameter restricted to the actual space available where a typical SLOWPOKE-2 reactor core currently resides, while still achieving criticality. The aim of this follow-on work is to conduct a safety analysis of the homogeneous design. Both deterministic and probabilistic modelling programs (WIMS-AECL and MCNP 5, respectively) will be employed to demonstrate the reactor's inherent safety. Historical data from an existing homogeneous reactor (ARGUS) will be used to validate the numerical models of the homogeneous SLOWPOKE reactor. Time-dependent modelling of heat transfer will be explored using COMSOL to ensure that natural convection is still a valid option for core cooling. Lastly, several modifications to the proposed design will be modelled or recommended, based on the results of the safety analysis.

INTRODUCTION

The thesis will be a continuation of the feasibility study performed by Lt(N) Paul Busatta[1]. Busatta's work focused on investigating the feasibility of replacing an existing SLOWPOKE-2 fuel assembly with a container filled with a homogeneous aqueous solution of Uranium Sulphate, mostly performed from a neutronics view point. The design reactor is intended mainly for the production of medical isotopes with possible side benefits of maintaining the ability to continue neutron activation analysis and other research activities currently performed by SLOWPOKE-2 facilities. The possible financial and educational benefits to the institutions operating SLOWPOKE-2 reactors make this a very attractive proposition.

This paper will discuss the development of a full safety analysis of the homogeneous SLOWPOKE design through computer modelling using both Winfrith Improved Multi-group Scheme – Atomic Energy of Canada Limited (WIMS-AECL) and Monte Carlo Neutron Particle-5 (MCNP-5) modelling programmes. Due to the lack of an actual homogeneous SLOWPOKE from which to obtain experimental data, validation of the experimental models will be accomplished through modelling of an existing homogeneous reactor design (ARGUS). Comparison of the simulated data with actual experimental data from this reactor can then be made.

AQUEOUS HOMOGENEOUS REACTORS GENERAL

Some of the main benefits of aqueous homogeneous reactors as they relate to research reactors[2] are summarized below:

a. High Specific Power. There are no heat transfer barriers between the fuel and the coolant/moderator. The greater mass of the core as a whole, as opposed to individual fuel elements in a heterogeneous reactor, can accept greater amounts of energy without overheating.

b. Continuous Removal of Fission Products. Online removal of fission products is possible, including Molybdenum-99 (Mo-99). Very high fuel burn-up and online refuelling are also possible, but less of a concern with the expected long lifetime of the fuel at the power levels contemplated, which are nearly identical to that of the ARGUS[3].

c. Simplified Reactor Core Design. The fuel and moderator (also acting as the core integral coolant) are one solution. This results in a relatively low cost of fuel preparation – and no requirement for fabricating of complex fuel bundles with engineered cladding based on expensive materials such as Zirconium.

d. High Neutron Economy. The absence of most internal structure (i.e.- cladding, structural) within the core drastically reduces parasitic neutron absorption.

e. Intrinsically High Negative Temperature Coefficient of Reactivity. The nature of the system is self-regulating such that an increase in the temperature leads to a decrease in the density of the aqueous solution, leading in turn to a reduction in the reactivity.

Liquid homogeneous research reactors were popular early in the development period of nuclear reactors, but only five were operating as of April 2006, as shown in Table 1.

Table 1. The World's Five Operating Aqueous Homogeneous Research Reactors as of April 2006 [4,5,6]

<table>
<tr><th>Serial</th><th>Reactor Name</th><th>Thermal Power</th><th>Location</th><th>Operator</th></tr>
<tr><td>1</td><td>SILENE</td><td>1.00 kW</td><td>Valduc, France</td><td>Commissariat à l'Énergie Atomique</td></tr>
<tr><td>2</td><td>ARGUS</td><td>20.0 kW</td><td rowspan="2">Moscow, Russia</td><td rowspan="2">Kurchatov Institute (Russian Research Center)</td></tr>
<tr><td>3</td><td>GIDRA</td><td>10.0 kW</td></tr>
<tr><td>4</td><td>IGRIK</td><td>30.0 kW</td><td rowspan="2">Snezhinsk, Russia</td><td rowspan="2">All-Russian Scientific Research Institute of Technical Physics (VNIITF)</td></tr>
<tr><td>5</td><td>YAGUAR</td><td>10.0 kW</td></tr>
</table>

The main technical reasons for the loss in popularity of the homogeneous reactor have to do with the following issues[7], which have been described as "daunting design and material challenges"[8]. Once again, the list is described in terms that apply to the relatively low temperatures of a research reactor:

a. Extreme Corrosiveness of the Fuel/Moderator Solutions. An example of early problems with the corrosiveness of fuel-moderator solutions is seen in the Oak Ridge National Laboratory's (ORNL) Homogeneous Reactor Test[8]. For this reason, Busatta strived to reduce the molality of the Uranyl Sulfate solution while maintaining acceptable levels of enrichment[9]. Advances in materials science[10] since the pioneering reactors have also provided new options for reactor materials, such as the Zircaloys.

b. Gas Formation through Water Dissociation. Water in an aqueous reactor appears to boil in conditions of high power output (hence historical name of water boilers for the first homogeneous reactors built at ORNL[8]). Although the effect is reduced at lower power levels, the relatively smaller amounts of the highly explosive mixture of hydrogen and oxygen gas must still be safely dealt with in the system.

c. Requirement for External Circulation of Fuel. To take advantage of the ability to refuel and eliminate fission products online, more fuel is required than the strict minimum that would be required to achieve criticality in the tank. The circulation of the Uranyl Sulphate solution outside of the reactor core also leads to a requirement for more shielding and possible maintenance problems due to the added complexity of the system and high radiation dose environments outside of the core.

PRODUCTION OF MOLYBDENUM-99

General. Mo-99 is produced as a precursor isotope for Technetium-99m, which is a widely used radioisotope in nuclear medicine. Standard production methods involve irradiating solid Uranium-235 targets with thermal neutrons, followed by chemical dissolution of the targets and extraction of the fission product Mo-99[11]. The standard process is complex, incomplete (in terms of fission of U-235), and produces significant radioactive waste[12].

Using a Homogeneous Reactor. In 1998, the Kurchatov Institute in Moscow developed an extraction method that removed Mo-99 from the ARGUS reactor solution through use of a proprietary sorbent material. The remaining solution, including the uranium, passed through and continued to assist with reactor operation. The Mo-99 was eluted from the sorbent with an acid solution and concentrated for further purification to meet medical standards[13]. The proposed homogeneous SLOWPOKE will make use of a very similar process.

RMC'S HOMOGENEOUS SLOWPOKE REACTOR

Parameters. Table 2 provides a summary of the characteristics of the proposed design. Limitations on the dimensions for the design were determined by the space constraints within standard heterogeneous SLOWPOKE-2 reactor reflectors. Figure 1 illustrates the concept of the proposed reactor designed at RMC.

Table 2: Summary of Homogeneous SLOWPOKE Reactor Characteristics[14]

Core height	48.8 cm
Core radius	10 cm
Core cladding thickness	3 mm
Cladding material	Zircaloy-4
Control rod orifice radius	0.73 cm
Control rod material and radius Control rod cladding thickness	Cadmium 2 mm Al 2 mm
Fuel volume at 313 K	15.244 L
Beryllium reflector annulus inner radius	11 cm
Beryllium reflector annulus outer radius	21 cm
Beryllium reflector annulus height	48.8 cm
Fuel	uranyl sulfate solution in water
Fuel enrichment	20%
Fuel concentration	1.65 M
U-235 Mass	1.181 kg
k_{eff}	1.00361
Thermal power	20 kW
Operating temperature at steady state	313 K

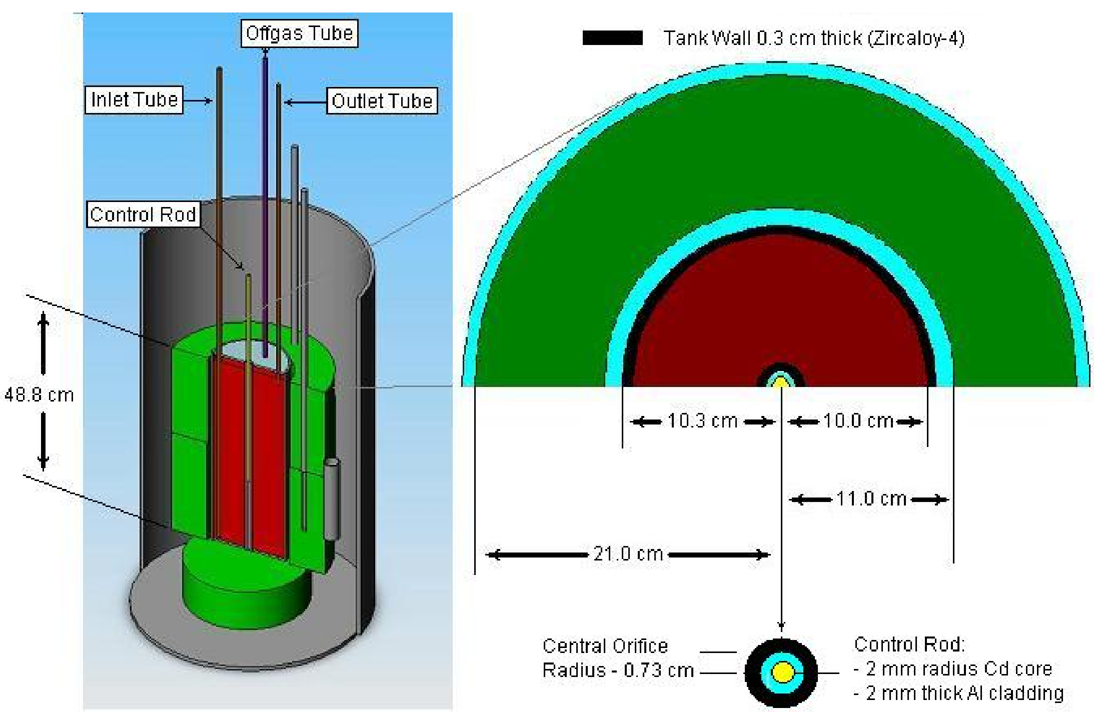

Figure 1. RMC's Reactor Concept (adapted from Reference [14,15])

Tank Materials. The results of a literature review conducted at Reference [16] determined that a dilute aqueous solution of Uranyl Sulphate (UO_2SO_4), H_2SO_4 and $CuSO_4$ with an oxygen overpressure was the fuel of choice for a small homogeneous reactor in terms of materials chemical compatibility with both Type 347 stainless steel and Zircaloy-2. Comparison of Zircaloy-2 and -4 elemental compositions and properties leads to the conclusion that Zircaloy-2 (as opposed to Busatta's proposed tank material[14]) is the more suitable material for the homogeneous SLOWPOKE tank, despite lesser relative resistance to hydrogen embrittlement[17].

Results of the Feasibility Study. The initial study determined that RMC's reactor design was feasible with the dimensional constraints imposed by typical SLOWPOKE-2 installations. The work demonstrated that the necessary conditions for inherent safety were met by the proposed design, with a strong negative reactivity coefficient due to temperature evidenced by the simulations. Initial calculations also determined that the single control rod was sufficient to shut down the homogeneous reactor, and that natural convection would provide sufficient cooling for the reactor tank while in steady-state operation[18].

PROPOSED SAFETY ANALYSIS

Approach. The following are the proposed steps in the safety analysis of RMC's Reactor, and any subsequent modifications:

a. Modelling of the ARGUS Reactor using MCNP-5 and WIMS-AECL. Using the same method as Paul Busatta, taking the output file of the MCNP-5 and modifying the buckling of the WIMS-AECL until the k_{eff} match is within tolerance.

b. Comparison of the Computed Data with Measured Data. The computed results will be compared to one another and to measured ARGUS data to confirm the validity of the approach. A convergence of the probabilistic and the deterministic models is also expected with the model of the ARGUS reactor.

c. Modelling of the Homogeneous SLOWPOKE using MCNP-5 and WIMS-AECL. This process is identical to Step 1. At this phase only a minor modification to Busatta's design will be implemented, involving a switch to Zircaloy-2 for the tank material[16].

d. Modelling of the Design Reactor using COMSOL. Time-dependent thermalhydraulics of the design will be assessed using COMSOL.

e. Modifications. Possible modifications as outlined below will be assessed singly or in combination through a repeat in steps c and d.

Possible Modifications. Further modifications to RMC's homogeneous SLOWPOKE design will be explored singly or in combination are listed below. Most modification concepts were first developed at Reference [19], unless otherwise indicated. These modifications will either be explored during the development of the analysis, or will serve as recommendations for further development of the design:

a. Reflector Material. Adding a graphite extension to the reflector as opposed to the proposed beryllium extension in the Busatta design in an attempt to reduce the cost (with due note to the galvanic interaction between the two reflector materials). The original beryllium reflector from the SLOWPOKE-2 reactor remains in place.

b. Inclusion of Extra Reflectors. The homogeneous SLOWPOKE design does not require regular addition of extra reflector shims as with the SLOWPOKE-2. The addition of a top reflector and completion of the bottom reflector would allow for a reduction in the solution molality.

c. Placement of Control Rod(s). Relocation of the control rod(s) to the reflector, possibly taking up one or more of the irradiation sites. This design alteration could lead to improved reliability and less complexity related to the reactor tank by removing the requirement for the central orifice. The gain in space will also allow an increase in the volume within the vessel and a possible consequent reduction in Busatta's proposed Uranyl Sulphate concentration.

d. Volumetric Control of Reactivity. Possible elimination of the need for a control rod through variation of the volume of Uranyl Sulphate Solution within the reactor tank (requiring extra piping and a subcritical holding tank). One negative aspect of this modification, added complexity aside, would be the extra volume of Uranyl Sulphate solution required.

e. Fuel Composition. Possible changes to the fuel composition include using thoriated fuel for purposes of breeding Uranium-233 [8], thus improving the reactor performance and possibly allowing a lower solution molality.

f. Heat Transfer Mechanisms. After analysis with COMSOL, any of the above modifications may result in the need for changes to the heat transfer mechanism employed.

CONCLUSION

The present work continues as a first step in the design of the homogeneous SLOWPOKE reactor, intended mainly for the production of radioisotopes for nuclear medicine. Validation of Busatta's approach in modelling the proposed design will be achieved using the same computer codes to model the ARGUS reactor. Time-dependent modelling of thermalhydraulics will further confirm that the natural convection cooling mechanism for the homogeneous SLOWPOKE core remains adequate. Several proposed design modifications will also be modelled or recommended based on results of the safety analysis. The promise of Mo-99 isotope production while maintaining research activities such as neutron activation analysis, all within existing SLOWPOKE-2 facilities, makes the homogeneous SLOWPOKE research reactor a concept worth pursuing.

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