HOMOGENEOUS SLOWPOKE REACTOR FOR THE PRODUCTION OF RADIO-ISOTOPE
A FEASIBILITY STUDY

Paul Busatta
Master Student
Royal Military College of Canada
Kingston, Ontario
Phone (613)541-6000 #6054
paul.busatta@rmc.ca

Hugues, W. Bonin
Professor
Royal Military College of Canada
Kingston, Ontario
Phone(613)541-6000 #6613
bonin-h@rmc.ca

ABSTRACT

The purpose of this research is to study the feasibility of replacing the actual heterogeneous fuel core of the present SLOWPOKE-2 by a reservoir containing a homogeneous fuel for the production of Mo-99. The study looked at three items: by using the MCNP Monte Carlo reactor calculation code, develop a series of parameters required for an homogeneous fuel and evaluate the uranyl sulfate concentration of the aqueous solution fuel in order to keep a similar excess reactivity; verify if the homogeneous reactor will retain its inherent safety attributes; and with the new dimensions and geometry of the fuel core, observe whether natural convection can still effectively cool the reactor using the modeling software FEMLAB®. It was found that it is indeed feasible to modify the SLOWPOKE-2 reactor for a homogeneous reactor using a solution of uranyl sulfate and water.

Molybdenum 99 Production Using a Homogeneous Reactor

Molybdenum 99 (Mo-99) is used to produce Technetium 99m (Tc-99m), the most widely used radioisotope in nuclear medicine. Its short half-life of six hours and its emitted gamma energy of 140 keV make it an ideal imaging agent. Mo-99 is obtained largely from the fission of U-235. Targets of U-235 are exposed to a neutron flux in a nuclear reactor**.** The target is an uranium/aluminium alloy, in which highly enriched uranium (HEU) or low enriched uranium (LEU) is used. The irradiated targets are then dissolved in an alkaline or acidic solution and the fission product of the U-235, Mo-99, is extracted by solvent extraction from the solution of targets and solvent by chemical treatment steps such as passing through several absorber columns and ion-exchangers. This method creates important amount of radioactive waste from the U-235 targets. Table 1 presents an overview of all the liquid produced for the production of 3000 Ci of ^{99}Mo in the Netherlands. Even though several methods are used by other manufacturers, this method still gives a good approximation of the waste produce[1].

Research has been conducted to reduce, handle, and treat the waste created by the production of Mo-99. In 1992, Russell M. Ball introduced the use of a homogeneous nuclear reactor for the production of Mo-99 with the following advantages[2] :

- No requirements to produce and transport targets;
- Full utilization of fission product Mo-99. In target irradiation, all the fission products (and the resulting production of Mo-99) in the reactor used to produce the neutrons that irradiate the target are wasted. The Mo-99 remains in the solid reactor fuel;
- Discarded fission products are 1/100th of the total fission products produced in the target method for a given quantity of Mo-99;
- The uranium consumption and the heat production are 1/100th of that produced in the target method;
- The extraction process is simplified with no subsequent uranium dissolution required.

Table 1: Characteristics of Liquid Waste
per Hot Cell after Production of 3000 Ci ^{99}Mo. (Ref 1.)

Liquid From	Volume (l)	Content (g l^{-1})	Activity (MBq l^{-1})
Cell 1 Intermediate level waste (ILW)	8,2	NaOH 240 Al 20 U 0.005	^{89}Sr 740 ^{90}Sr 630 ^{137}Cs 6400
Cell 2 ILW	10.5	$NaNO_3$ 102 NaOH 29.6	^{103}Ru 500 ^{106}Ru 46 ^{125}Sb 4.6
Cell 3 Low Level Waste (LLW)	7.3	Na_2SO_4 115 NaI 0.06 Na_2SO_3 121 NaOH 16 NaSCN 4	^{103}Ru 5 ^{106}Ru 0.46 ^{125}Sb 0.046
Cells 4 and 5	4.7	NaOH 12 $NaNO_3$ 17	^{103}Ru 0.040 ^{106}Ru 0.0037

In 1998, the Kurchatov Institute designed a Mo-99 extraction procedure using a homogeneous reactor. The irradiated fuel solution is then passed through a proprietary sorbent, which separates the Mo-99 from the rest of the solution, the remaining solution being sent back to the reactor. The Mo-99 is then separated from the sorbent using an acid solution.

<u>Homogeneous Nuclear Reactor</u>

Homogeneous nuclear reactors are reactors for which the fuel, the coolant and the moderator are distributed uniformly and homogeneously throughout the core. The fuel and moderator could be solid or liquid (the coolant must be a fluid (liquid or gas)). The type of homogeneous reactor of interest for this study is the aqueous homogeneous reactor. The aqueous homogeneous reactor uses the fuel in an ionic state in an aqueous solvent that is used as coolant as well as and moderator. In contrast, in the heterogeneous nuclear reactor, the fuel is kept physically separated from the moderator (and coolant) by means of cladding (or sheathing). The following is a list of all the advantages and disadvantages of the homogeneous reactor in the liquid aqueous form.

Advantages of homogeneous nuclear reactors.

1. Reactor's high specific power. Because the coolant and fuel are part of the same solution, there are no heat transfer barriers and the power density is only limited by the temperature rise of the fuel itself and the heat removal capacity of the system.
2. High burn-up of fuel. the aqueous homogeneous reactor has the possibility of removing the so-called "poisons fission products" and adding fresh fuel on a continuous basis, therefore permitting very high burn-up and very efficient use of the nuclear fuel.
3. Simple fuel preparation and processing. There is no need of complex and expensive fuel element fabrication, resulting in lower costs of fuel preparation for the homogeneous reactor.
4. Loading and discharging of fuel can be done on-line, thanks to a extensive fuel handling facility, which constitutes yet another advantage.

5. Homogeneous reactors allow high neutron economy in minimizing the neutron absorption by avoiding the need for cladding and structural material within the reactor core.
6. Simple control systems are possible with homogeneous reactors. Increasing the core temperature leads to a decrease of the homogeneous fuel mixture density, thus creating a strong negative temperature coefficient of reactivity which makes the system self – stabilizing, thus eliminating the need for complicated safety systems.[3]

Disadvantages

1. Corrosion or erosion of equipment: The circulation of the homogeneous fuel mixture at high flow rates creates corrosion and erosion problems in the reactor and circulating equipment.
2. The external circulation of the fuel solution: In order to remove the fission products and isotopes from the fuel and to condition the fuel outside the reactor core, the highly radioactive mixture is circulated outside the reactor core.
3. The inventory of the fuel: It needs to be higher than the content of the volume of the core. This increase of fuel inventory of fuel and the need to handle relatively large quantities of highly radioactive fuel mixture result in increased induced activity within in the external equipment and in additional shielding requirements and maintenance problems.
4. Restriction in the concentration of uranium in the fuel solution: In aqueous homogeneous reactors, the concentration of uranium is limited by solubility and the corrosion effects of high concentrated solutions. Figure 1, which presents the phase diagram for the system UO_2SO_4-H_2O, displays the level of solubility of UO_2SO_4-H_2O in moles per kg of water at different temperatures. As observed in Figure 1, the fuel solution becomes saturated at 4.3 moles of UO_2SO_4-H_2O per kg of water at a temperature of 50^oC.
5. The last disadvantage of the homogeneous reactor is the production of explosive gases. Radiation induces the decomposition of the moderator (water), which can produce an explosive mixture of hydrogen and oxygen within the reactor system. This hazard means that special precautionary design measures must be taken such the use of catalytic recombiners.[3]

Even though the list of advantages and disadvantages applies primarily to power generation reactors that require higher neutron fluxes, temperatures, pressures and coolant flow rates, it is still valid for smaller reactors albeit to a lesser extent, and the disadvantages may not represent obstacles important enough to preclude the design of a small homogeneous reactor such as the one investigated in the present work

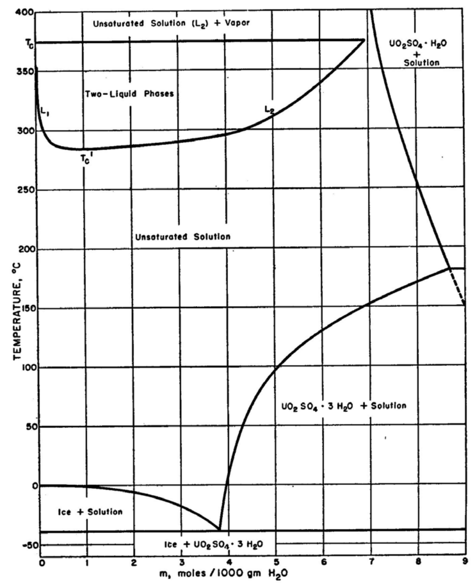

FIG. 1. Phase diagram for the system UO_2SO_4-H_2O.

Figure 1: Phase Diagram for the System UO_2SO_4–H_2O [Ref 3]

Goal of the Research.

Since some of the SLOWPOKE-2 reactors in service in Canada need to be refuelled in a near future, it would be interesting to study the feasibility of replacing the actual heterogeneous fuel core of the present SLOWPOKE-2 by a reservoir containing a homogeneous fuel for the production of Mo-99.

The study will look at the following items:

1. By using the MCNP 5[4] probabilistic nuclear reactor physics code, develop a series of parameters required for an homogeneous fuel and evaluate the uranyl sulfate concentration of the aqueous solution fuel in order to keep a similar excess reactivity;
2. verify if the homogeneous reactor will retain its inherent safety attributes; and
3. with the new dimensions and geometry of the fuel core, observe whether the natural convection will still effectively cool the reactor using the modeling software based on the finite element method FEMLAB®[5], or not.

The results obtained will be the stepping-stone for further development of a Homogeneous SLOWPOKE Reactor for research purpose and/or production of commercial radioisotopes.

The SLOWPOKE-2 Reactor

The Safe Low-Power "Kritical" Experiment (SLOWPOKE) reactor was developed by Atomic Energy of Canada Ltd (AECL) in the late 1960's. This reactor is a low power pool-type reactor, and is inherently safe. Because of its low excess reactivity and large negative temperature and void coefficients, it is licensed in Canada for unattended operation while remotely monitored without the need to have its operators extensively trained. The safety features can be summarized in the negative effect on the reactivity of a temperature increase or of a loss of its coolant/moderator (water). Figure 2 displays the geometrical arrangement of the SLOWPOKE-2 reactor.

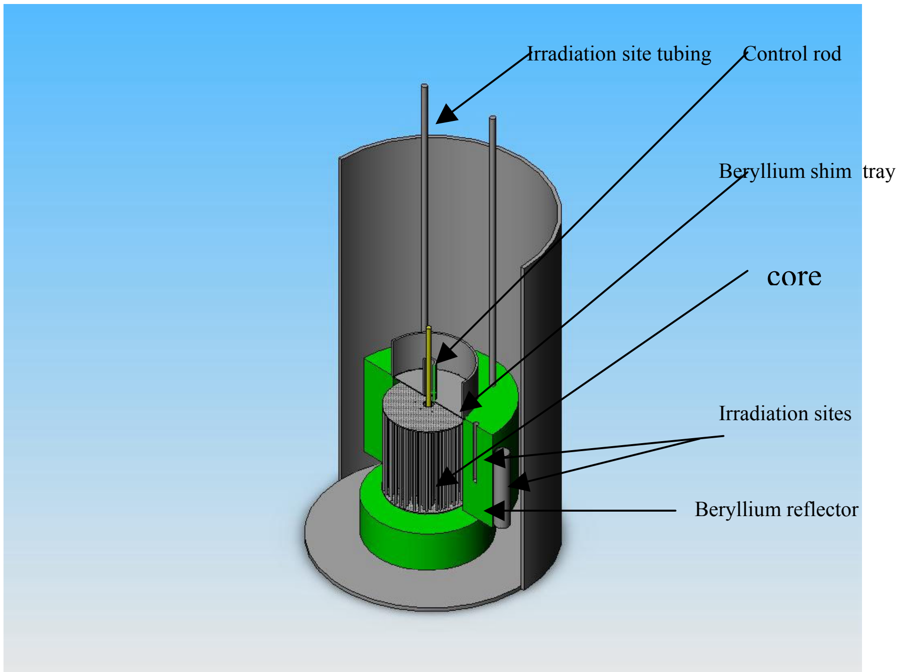

Figure 2: Simple Representation of the SLOWPOKE –2 Reactor.

<u>Homogeneous SLOWPOKE Reactor.</u>

In order to evaluate the feasibility of transforming the SLOWPOKE –2 reactor into a homogeneous reactor, several changes need to be made to the present design to allow the introduction of a homogeneous fuel without modifying the main structure and the original annular beryllium reflector. The changes being investigated are as follows and can be seen in Figure 3:

a. removal of the beryllium shims and tray;
b. removal of the actual fuel core and control rod;
c. insertion of a fuel core with homogeneous fuel. The core is made of a Zircaloy-4 container with an orifice in the centre vertical axis for the insertion of a control rod and cooling. The core is filled with an aqueous solution of uranyl sulfate. It is possible to extend the core height above the 22 cm height of the present core.
d. possibility of increasing the height of the beryllium reflector to follow the height of the core by adding an addition to the top of the annular reflector if the homogeneous reactor core is taller than the original core; and
e. modification of the control rod to provide sufficient negative reactivity to shut down the reactor at all times.

<u>Modeling of Reactor</u>

First an MCNP 5 model was constructed, to determine if the homogeneous reactor will have a positive excess reactivity without modifying the actual architecture of the reactor. To do this, so the actual core is replaced with a container made of Zyrcaloy-4 filled with a homogeneous fuel composed of a solution of water and uranyl sulfate. Several simulations were conducted by varying the volume of the container by changing its height to a maximum of 50 cm, which is dictated by the height of the lower core container (about 50 cm of height for the core),and the concentration of the solution to a maximum of 4 M of uranyl sulfate. The height of the beryllium reflector followed that the container. During the process, the concentrations of the solution were kept to a minimum to avoid corrosion of the container.

When the size of the reactor and the concentration of the fuel solution were established, the parameters were fine tuned to allow the addition of the control rod and the fuel treatment piping. Table 2 displays a summary of the final design. To validate the results, a model was constructed with the code WIMS-AECL[6] with the dimensions at Table 2. By adjusting the axial buckling, the results obtained with the 2-D representation by WIMS-AECL were similar to the results obtained with 3-D representation by MCNP-5.

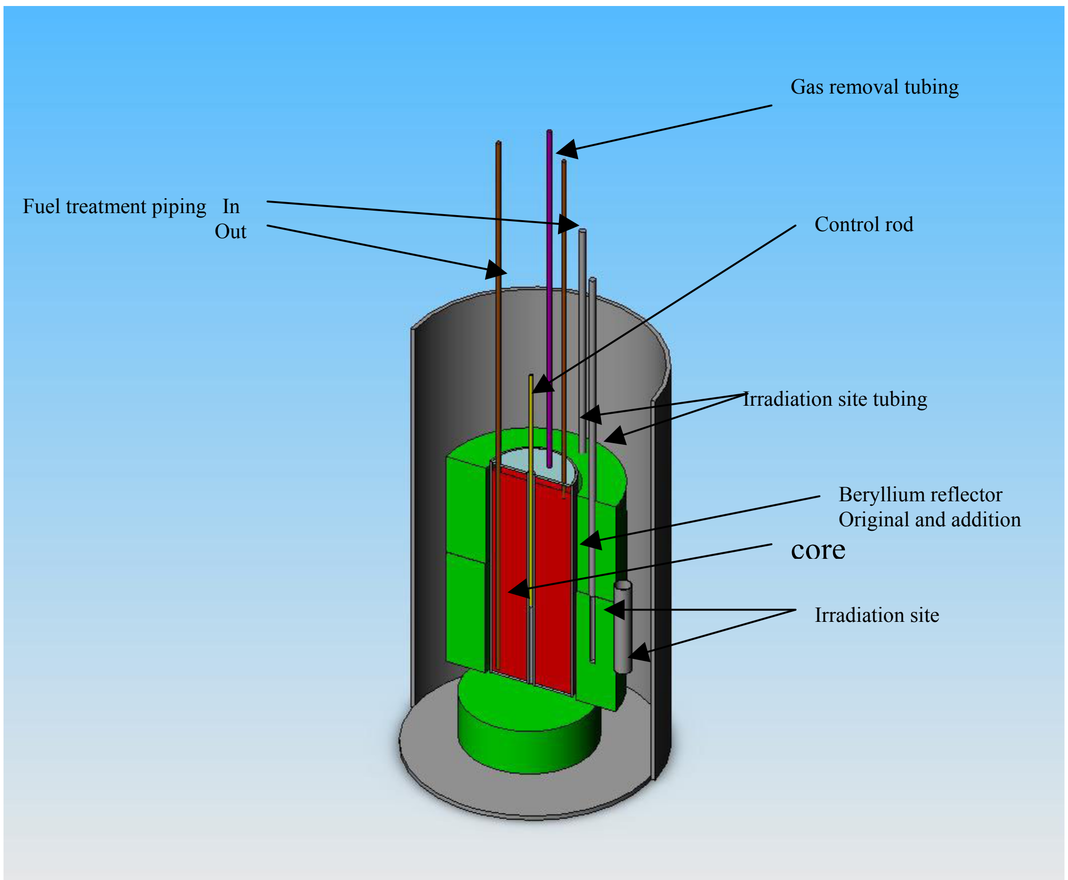

Figure 3: Simple Representation of the Homogeneous SLOWPOKE Reactor

Table 2: Summary of Reactor Characteristic

Reactor Parameters	
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 40^{o}C	15.244 l
Beryllium reflector annulus inner and outer radius	11 cm / 21 cm
Beryllium reflector annulus height	48.8 cm
Fuel	Uranyl Sulfate solution in water
Fuel enrichment	20%
Fuel Concentration	1.65 M
k_{eff}	1.00361 MCNP 5 result
Thermal power	20 kW
Operating temperature at steady state	313 K

<u>Effects of Temperature and Control Rod</u>

With the concentration and dimensions listed in Table 2, the inherent safety of the reactor was verified. Simulations were conducted by varying the temperature of the reactor using both MCNP-5 and WIMS-AECL. Figure 4 displays the effects of the temperature on the excess reactivity of the reactor. The results support the safe characteristic of the homogeneous reactor and that the reactor has indeed a negative reactivity factor at high temperature.

With the same method at the operating temperature, the effects of the control rod position were simulated with MCNP-5. Figure 5 displays these results. The results demonstrate that the use of the control can indeed shut down and control the reactor effectively.

<u>Cooling of the Reactor</u>

Since the reactor core design has changed significantly, a heat transfer study was conducted to evaluate if natural convection alone will be sufficient to cool the reactor. The study was conducted using the finite element based modeling software FEMLAB®. To save on computing time, the model was transposed into a two dimensions model, where the non-isothermal flow equation (variation of the Navier-Stokes equation) is coupled with the energy balance equation combined with the physical properties of each of the materials.

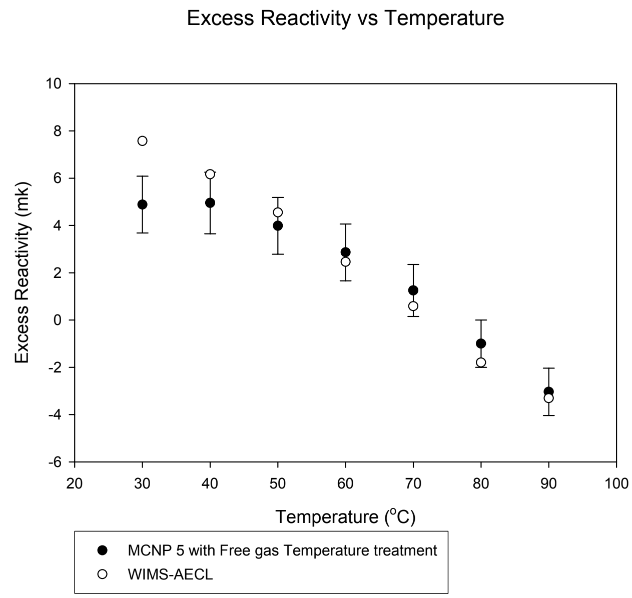

Figure 4: Effect of Temperature on Reactor Excess Reactivity using AECL- WIMS and MCNP-5

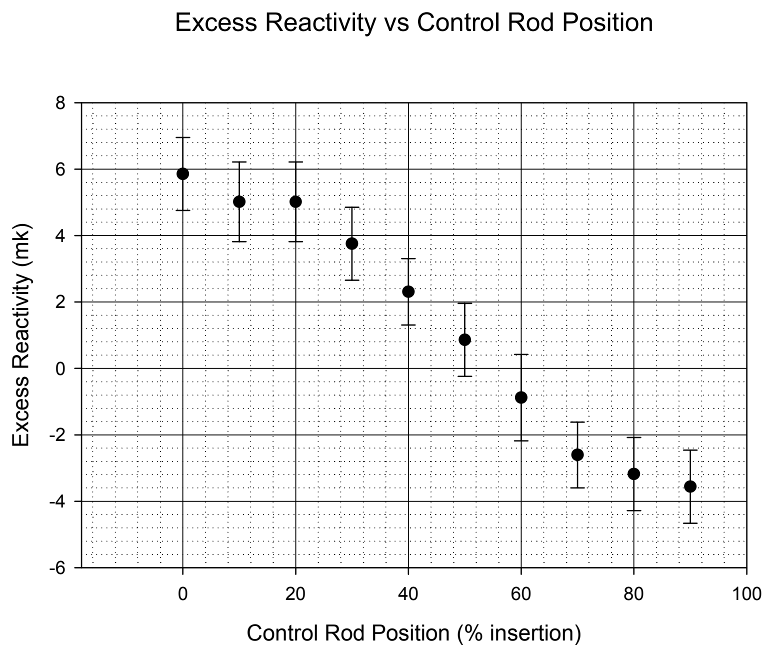

Figure 5: Excess Reactivity vs Control Rod Position using MCNP 5

Figure 6 presents the temperature distribution inside of an axial slice of the fuelled core at steady state from the centre of the reactor (centre of the control rod orifice) to the reflector (refer to Figure 3). The results show that natural convection is sufficient to cool the reactor to a temperature similar to that of the SLOWPOKE-2 reactor operating at full power, which is 315 K.

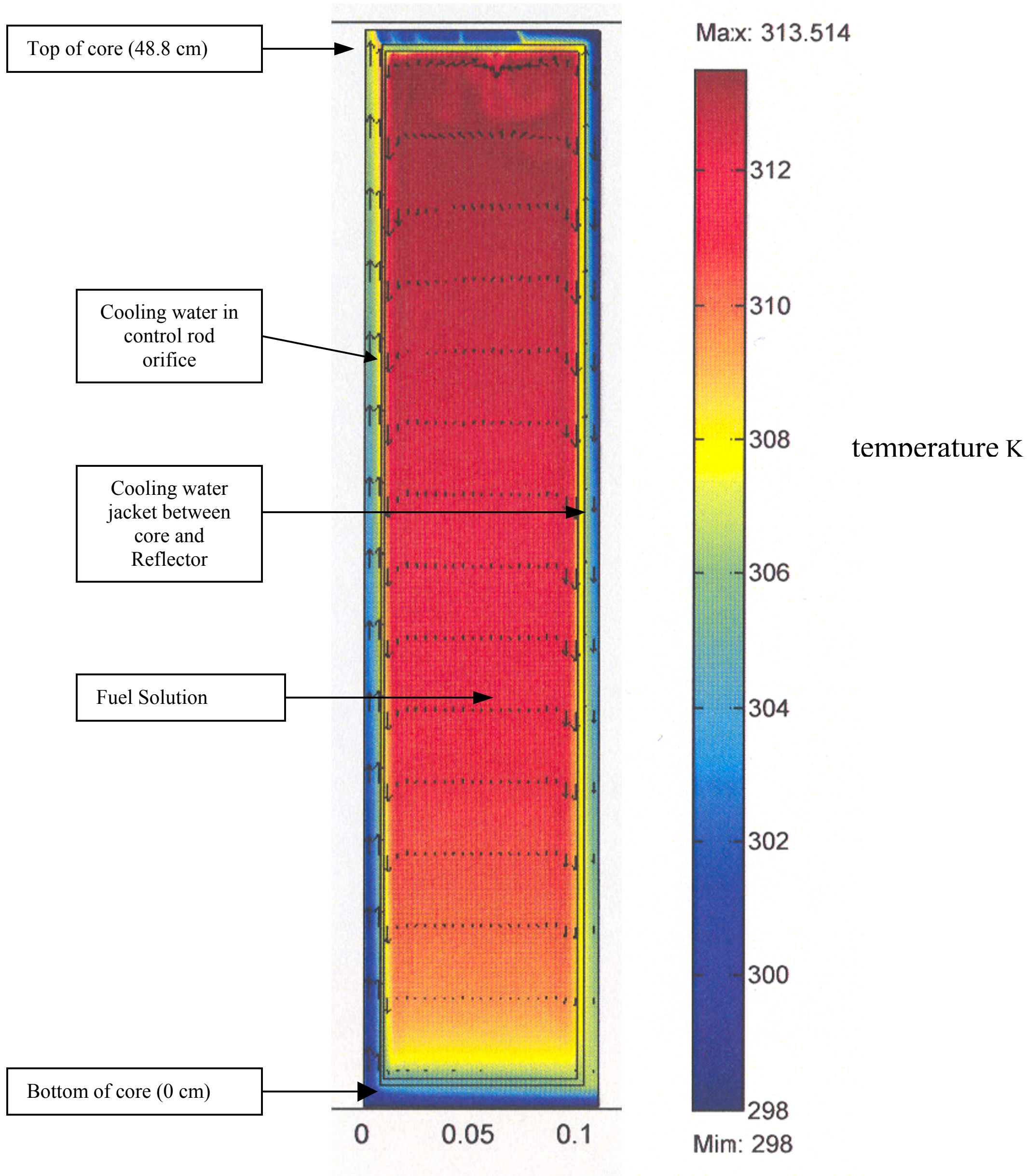

Figure 6: Flow Pattern and Temperature Distribution
of Fuel Mixture and Cooling Water at Steady State using FEMLAB®

Discussion of Results.

Neutronics Results: It is impossible to verify the results obtained from the simulations to corresponding values obtained experimentally, since no homogeneous SLOWPOKE reactors have been built and operated, the study had to use two different modeling codes, MCNP 5 and WIMS-AECL. MCNP 5 is a probabilistic code that solves the neutron transport equation using the Monte Carlo method for a three dimensions model, and WIMS-AECL is a deterministic code that solves the transport equation in two dimensions using the collision probability method. The legitimacy of the validation of the model with those two codes is improved by the similar geometry description provided as of the input file of both codes. This is due to the simplicity of the reactor architecture, which is composed of a series of concentric cylinder of different material as pictured in Figure 3.

MCNP-5 is a reliable code. Pierre[7] successfully modelled the present SLOWPOKE-2 reactor using MCNP-4, and compared his result to commissioning results of the RMC's SLOWPOKE-2 reactor. The simulation, even with a cruder version of MCNP, proved to be faithful to the actual reactor performance. Since the Homogeneous Reactor is simpler to be modelled due to a simpler geometry, and since a more advanced version of MCNP was used, the confidence on the result precision has increased.

Past work from Cole[8] and Lamarre[9] have established an error $\pm$ 5% when WIMS-AECL is used. Since WIMS-AECL code is based on a two-dimensional representation, an additional $\pm$1% is added for not explicitly accounting the third dimension. The error comes from three sources: one source of error comes, as mentioned, from the two dimensional model that simulates a three dimensional reactor. The second source of error comes from the fact that WIMS-AECL is a lattice cell code, in which one cell of an infinite number of cells in the reactor is represented by the code. Because the Homogeneous SLOWPOKE is built as one cell, the reflective boundary condition at the limit of the cell is changed from a mirror-type boundary to that of a non-reflective "void" boundary condition to allow for those neutrons to escape without any possibilities of back scattering and re-entering the reactor. The last source of error comes from the assumptions made in the codes, from two correction methods: one correcting the diffusion coefficient using the Benoist method and the other accounting for the neutron leakage by determining the radial and axial buckling.

In this case, the axial buckling of WIMS-AECL code was artificially adjusted such that the criticality results (ie k_{eff}) obtained by WIMS-AECL matches the value of MCNP-5. Then the temperature effects of both code were compared (Figure 4), and since the trends of both curves are similar, one can conclude that both codes appear accurate in modelling the homogeneous SLOWPOKE reactor.

<u>Heat Transfer Model</u>

The uncertainty of the results provided by FEMLAB® depends on the precision of the equations used as input and the tolerances set in the problem solving method. For the heat transfer analysis of the reactor at steady state, a relative tolerance of 0.1% and absolute tolerance of 0.01% were used. Therefore the error on the results originates from two possible causes. First, like the case of the other software used to analyse the criticality of the reactor, the geometry of the problem represents a source of the error. As for MCNP 5 and WIMS-AECL, the reactor geometry is simple to reproduce with the graphic user interface; therefore the error coming from the construction is considered small. Lastly, the error may come from the input data and equations. The majority of the parameters and equations used in the FEMLAB problem definition come from reliable data sources that were derived mostly from credible experimental studies. The only data that had to be approximated was the equation of the influence of temperature on the viscosity of uranyl sulfate, where the equation was curved fitted using only three data points. A parametric study was carried out to evaluate the sensitivity of the results on the values of the equation's coefficients, and confirmed the reliability of the FEMLAB® calculations.

Conclusions:

The aim of this work was to study the feasibility of converting a SLOWPOKE-2 reactor into a homogeneous inherently safe reactor. By using a numerical model of the reactor with the code MCNP 5, it was possible to reach criticality for the homogeneous reactor by increasing the height of the reactor. It was found that the concentration would reach saturation point if the original height of 22 cm were used, therefore a height of 48.8 cm was determined as necessary to keep the concentration to a minimum value to avoid corrosion problems and yet obtain a reactor with sufficient positive excess reactivity. Therefore, for a height of 48.8 cm, the concentration would vary between 1.6 M and 1.65 M for a resulting excess reactivity between 1 and 6 mk.

In this work, the necessary condition for inherent safety of the reactor was verified and confirmed since an increase of the temperature has indeed a negative effect on the reactivity such that the reactor would become sub-critical at a temperature of 75 ^{o}C . In addition, it was determined that a single control rod was sufficient to shut down the reactor by itself, without the need of additional shutdown system.

Lastly, a heat transfer study was conducted to confirm that natural convection would be able to cool the reactor down enough to avoid localized boiling of the fuel mixture and cooling water. It was found, using the simulation program FEMLAB®, that the fuel would reach a maximum temperature of 40 ^{o}C at steady state, which is well below the atmospheric boiling points of both the cooling water and the fuel solution.

With the change of its fuel, an increase of its core height and reflector the reactor and the addition of few ancillary equipment such as an extension of the annular beryllium reflector, it is indeed feasible to modify the SLOWPOKE-2 reactor into a homogeneous reactor using a solution of uranyl sulfate and water.

It is recommended the research continue with a emphasis on the on the safety of the reactor during power transients, the control system to be used and on the material to be used for the reflector addition.

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