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MEDICAL ISOTOPE SHORTAGE 2009 – 2010 & FUTURE OPTIONS NRU, SLOWPOKE & MAPLE

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Abstract – The 15 month shutdown of NRU and the unexpected termination of the AECL/Nordion MAPLE project caused a world-wide shortage of medical isotopes. After the recent repair of NRU, AECL is confident that it could continue operating safely and reliably as a multi-purpose reactor until 2021 or longer. There is convincing evidence that the restoration of the MAPLE reactors is technically feasible, but it is highly improbable that a 10 MW MAPLE production reactor can ever be cost-effective. However, conversion of the present 10 MW reactors to 3 MW, without major changes to the structural hardware, warrants serious consideration. Finally, even the 20 kW SLOWPOKE reactor could produce useful quantities of Mo-99. If the present fuel rods were replaced with a small tank containing a solution of low-enriched uranyl sulphate in water, three of these liquid core reactors could supply all of Canada.

1. NRU shutdown, 2009 – 2010

A global shortage of medical isotopes was caused by the unexpected outage of the NRU reactor from May 2009 to August 2010. [1] The discovery and repair of a heavy water leak at the bottom of the reactor vessel was unusually difficult because of limited access; and though costly, the successful repair was a remarkable technical achievement. AECL staff will continue to inspect the leak area, and they are confident that NRU can continue operating until 2021 and possibly much longer. However, another long shutdown, or permanent closure, would have a serious impact on the supply of medical isotopes to North America. [1] In particular, it would limit diagnostic testing of cancer patients using the isotope Tc-99m, derived from the radioactive isotope Mo-99; and Tc-99m cannot be stockpiled, because it decays with a half-life of 6.0 hours. The global demand for Tc-99m is approximately 40 million doses per year, of which 30% to 40% is normally supplied by NRU. [1] At an estimated price of $20 per dose, total annual sales would be $800 million. Figure 1 shows how NRU supplies North America and the rest of the world with Mo-99/Tc-99m.

Since 1957, the multi-purpose NRU reactor has been a unique national asset, serving Canada in the following fields:

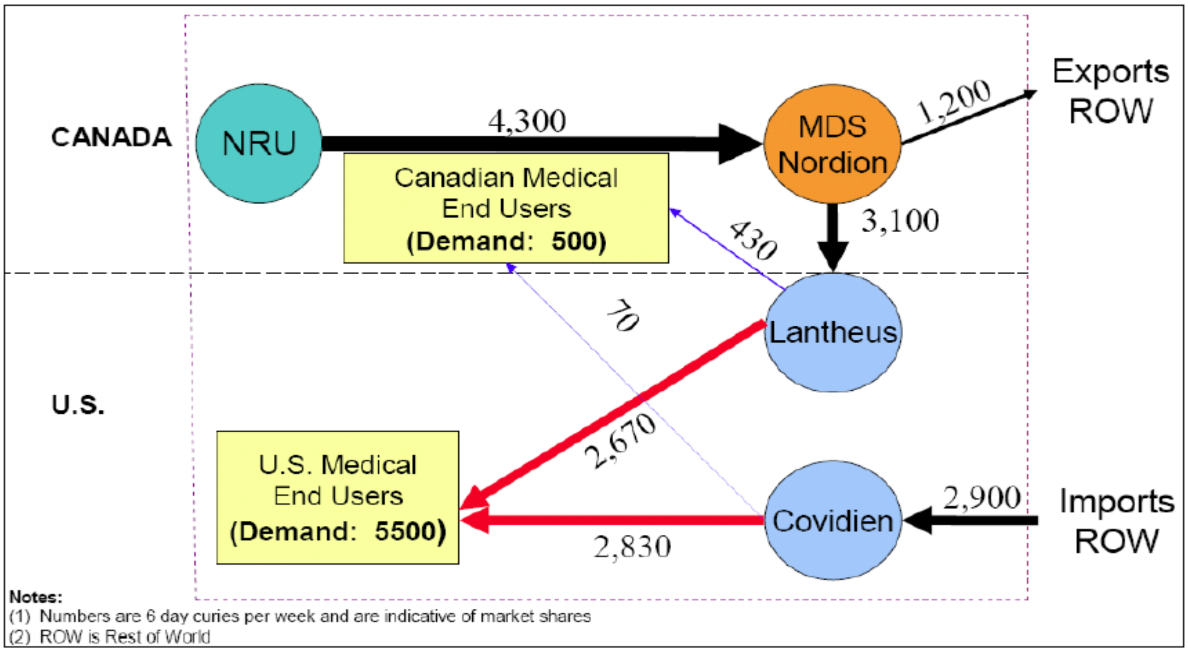
- Production of industrial and medical isotopes for an expanding world market. Before the 2009 shutdown, medical isotopes from NRU were helping more than 76,000 people daily, in over 80 countries. [1]
- Large-scale facilities for testing CANDU reactor materials and fuel assemblies.
- Neutron beam research, advancing the science of solid state physics, both applied and fundamental. (In 1994 Bertram Brockhouse was awarded a Nobel Prize for his “pioneering development of neutron scattering techniques for studies of condensed matter”, using a high-flux neutron beam from the NRU reactor.)

- Outreach program. NRU's versatile research facilities receive over 200 professors, students and industrial researchers annually. [1]
- With no replacement reactor planned in Canada, there is no technical reason why NRU could not be restored to service for another 20 years. A few outstanding licensing issues would have to be resolved, and ultimately it may become necessary to replace the reactor vessel. Since the vessel has already been replaced once before, the required technical knowledge, skills and documentation are all readily available. Many of us with extensive NRU operating experience are confident that it is technically feasible, and we also believe that it would be prudent to order a new vessel now, even if it is not required in the near future. World wide, NRU is still among the top five high-flux, multi-purpose research reactors.

As noted by former NRU Manager Don Ross in a letter to the North Renfrew Times (September 8, 2010*): "Congratulations to the team that repaired and restored NRU to operation. However it should be recognized that it was not a unique situation. In over 50 years of association with NRU, I have seen repairs to the original reactor under the same difficult conditions. As the corrosion continued, it was necessary to remove the vessel and install the replacement vessel. In the late 70's, while in charge of NRU, I was told that NRU could not possibly last until 1990. It is still going strong. ……Now is the time to build a replacement reactor vessel for NRU. This will cost far less than a new reactor. The corrosion will continue to create new leaks and in future years another crisis will occur. AECL should not drift into inaction and miss the opportunity. Canadians deserve better."*

If a new vessel is installed, there could be an added bonus. By filling the J-rod annulus with graphite or beryllium, it may be possible to increase the ratio of neutron flux to power. That is, the neutron flux could be increased while NRU continued to operate at the current level of 100 MW. The reflector annulus surrounding the reactor core reduces neutron leakage, thereby reducing the required loading of uranium fuel to achieve criticality; and in a thermal reactor the ratio of thermal neutron flux to fission power is approximately proportional to the inverse of U-235 mass in the core.

Figure 1 –World Distribution of Mo-99 from NRU [1]



Source: Natural Resources Canada, Document presented to the House of Commons Standing Committee on Natural Resources, Meeting 23, 2 June 2009.

2. Homogeneous SLOWPOKE reactor

Research at the Royal Military College in Kingston indicates that the present 20 kW SLOWPOKE 2 reactor could be converted to homogeneous operation by replacing the present core inside the beryllium reflector with a 15 litre tank containing a solution of uranyl sulphate in water. [2] The uranium would be enriched to 20% U-235, designated LEU (**L**ow **E**nriched **U**ranium).

Three 20 kW homogeneous reactors or two 30 kW* reactors with daily extraction of Mo-99 from the whole core, and distribution to regional hospitals within two days, could meet the Canadian demand of approximately 500 six-day Curies per week. [Fig. 1] The six-day Curie is the commercial unit of radioactivity for Mo-99; and it equals reactor Curies reduced by six days of radioactive decay, a factor of 0.2205. Fission reactors produce 51Curies of Mo-99 per kW at equilibrium (~11 days), and after 23 hours the radioactivity has reached 21.5% of equilibrium, or 11 Curies/kW. Assuming two days for chemical processing and distribution of Mo-99 generators, and 10% losses in extraction of Mo-99, the output from one 20 kW homogeneous SLOWPOKE reactor would be 185 six-day Curies per week.

An important advantage of the homogeneous SLOWPOKE is that after daily removal of the active solution from the 15 litre tank, it can be refilled immediately from a second tank during a reactor shutdown of less than an hour. After hot-cell extraction of the Mo-99, the active fuel solution can be recycled the following day. However, at periodic intervals it will be necessary to separate, solidify and encapsulate the waste fission products, prior to long-term storage.

From the beginning, inherent safety has been a cornerstone of the SLOWPOKE concept, and the 20 kW Homogeneous SLOWPOKE will incorporate the following safety and control principles:

- Maximum excess reactivity less than prompt critical by design.
- Negative temperature and void coefficients.
- Triple containment of the water-cooled core,
- Natural convection cooling of the core vessel wall at atmospheric pressure.
- Core vessel sub-critical in water, critical only when inserted inside beryllium reflector.
- No control rods or cooling tubes inside the reactor vessel.
- Two motor-driven control rods in the beryllium reflector.
- Independent manual shutdown system; fast-response automatic system not required.
- Control of neutron flux over two decades with one self-powered neutron flux detector.
- Start-up from cold shutdown in less than 5 minutes, no special instrumentation required.

In 1970, transient tests at Chalk River demonstrated that loss-of-control accidents approaching prompt-critical are safely limited by the negative reactivity coefficients. A rapid removal of the control rod resulted in a power peak of 180 kW and a temperature peak of 95 degrees C. [3]

In 1988, chemical extraction of Mo-99 from a homogeneous reactor in Taiwan reported 81.0% recovery of Mo-99 and 98.5% recovery of uranium. The fuel solution was 183g LEU/litre uranyl sulphate in a 0.3 molar sulphuric acid solution. [4]

* The Chinese “SLOWPOKE” reactor is rated at 30 kW.

3. MAPLE reactor termination

Following an agreement between AECL and Nordion in 1996, two isotope production reactors were constructed at Chalk River. Designated MAPLE 1 and MAPLE 2, the two reactors were completed in May 2000. However testing of MAPLE 1 revealed a positive power coefficient of reactivity (PCR), which was not predicted by the mathematical model. This discrepancy had a serious impact on safety and licensing, and despite intense efforts to rectify the problem, no practical solution was found. Consequently AECL terminated the project in May 2008, and Nordion claimed compensation. Subsequently the two parties commenced Arbitration by a Tribunal, and in September 2012, after three years of arbitration, AECL reported that a majority of the Tribunal affirmed AECL’s position and dismissed Nordion’s arbitration claim.

When the PCR discrepancy was confirmed beyond doubt, a number of solutions were proposed, but in June 2009 former AECL President Hugh MacDiarmid was quoted as saying it would take “many years and hundreds of millions of dollars before (they) would be licensable and could be put into service.” [1] Having studied the general problem of supplying Tc-99m/Mo-99 and other medical isotopes to Canada and the world for the past three years, I have come to the conclusion that it is extremely unlikely that any 10 MW reactor dedicated to commercial isotope production could possibly be cost effective, at isotope prices that hospitals in Canada and elsewhere could afford in the foreseeable future.

Several years ago South Africa planned to construct a new 10 MW reactor dedicated to commercial isotope production, but in March this year decided to expand its feasibility study to include a multi-purpose reactor for isotope production, nuclear fuel testing and research. [5] With that broader mandate, the cost of building and operating the reactor could be shared between isotope production and research, as is the case for NRU and all other major isotope suppliers.

The present proposal for modifying the MAPLE reactor is to replace the 10 MW core with a 3 MW beryllium-reflected SLOWPOKE reactor, utilizing the existing 19 channel hexagonal geometry and structural hardware. Let's call it MAPLE-SP. Fortuitously, the total cross-section area of the seven central channels of the MAPLE core is almost the same as the cross-section area of the SLOWPOKE 2 cylindrical core, inside the beryllium reflector. [Figures 2 & 3]

All of the present driver fuel would be eliminated, and the 7 central channels would be loaded with annular LEU targets of similar design to the present HEU MAPLE targets, but with a thicker annulus of uranium oxide. The 12 outer channels would be loaded with water-cooled beryllium blocks, 6 hexagonal and 6 cylindrical, as in the existing MAPLE layout. The present control absorbers and safety absorbers would be retained, and the heavy water reflector and irradiation sites would also remain as is. In order to guarantee that the power coefficient of reactivity is negative, the H/U-235 atom ratio in the central channels would be adjusted to the same value as in SLOWPOKE-2 fuelled with LEU: and we expect the water-cooled beryllium channels to have a negative temperature coefficient of reactivity.

Preliminary Monte Carlo calculations (MCNP, Los Alamos) indicate that less than 2 kg of U-235 is required in the 7 fuel/target channels of MAPLE-SP. This is possible because of the remarkable neutron economy of a small diameter HEU/water core with a beryllium reflector. [6] Therefore it is both feasible and beneficial to eliminate the wasteful driver fuel and utilize the entire reactor core for Mo-99 production. The excess reactivity is sufficient to compensate for xenon, samarium, the negative power coefficient, fuel burn-up and a short shutdown. With no driver fuel and weekly replacement of the entire core, the fuel burn-up reactivity is minimal, the in-core inventory of hazardous long-lived fission products is minimal, and the cost of fabricating driver fuel, and storing the driver fuel waste is entirely eliminated.

With the 7 central channels each operating at an average power of 430 kW, the reactor power would be 3 MW. Assuming that one channel is removed per day at 83 % equilibrium irradiation, the daily Mo-99 production from one MAPLE-SP reactor would be 1.4 times the daily production from NRU, which has been providing approximately 40% of world demand. Therefore assuming the same annual capacity factor for both reactors, **one 3 MW MAPLE-SP reactor could provide more than half of the world demand.**

Figure 2 MAPLE Core Layout

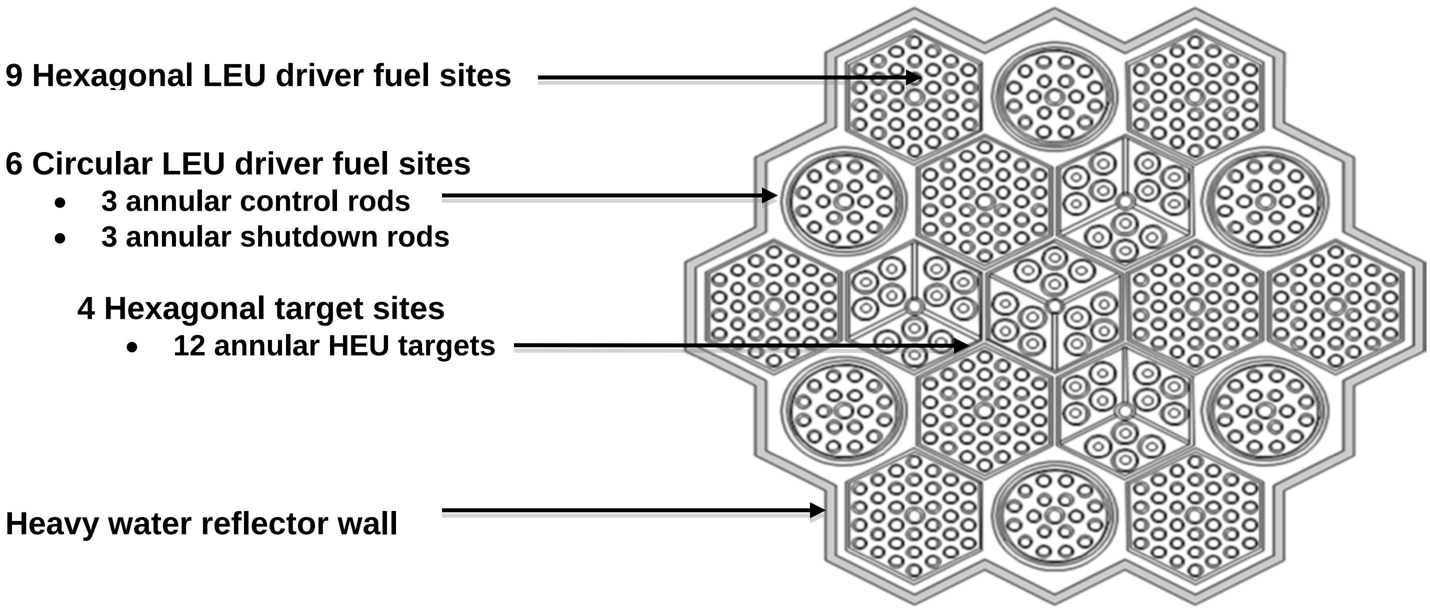
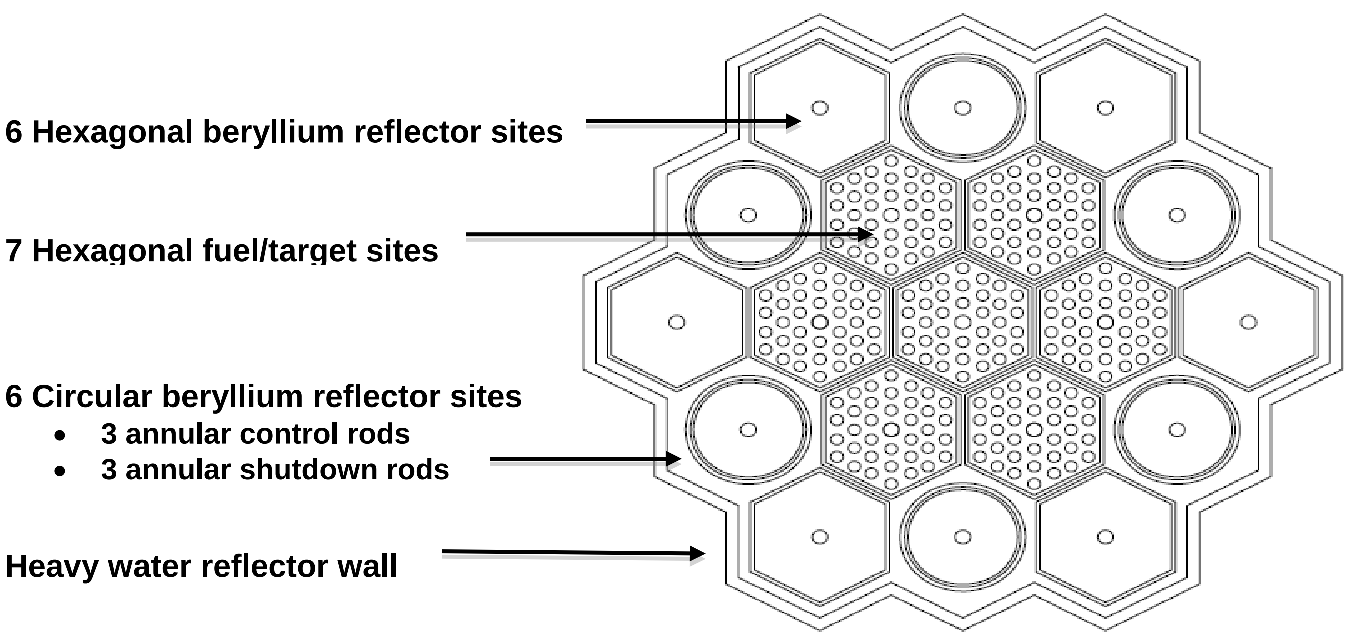


Figure 3 MAPLE-SP Core Layout



3.1 Fuel/target options

Although the original MAPLE targets use HEU, MAPLE-SP fuel/targets would probably be required to use LEU. However, since HEU might be authorized on a short-term basis for test purposes, it is included among the options listed below.

- Four, six or seven fuel/target sites, with the remainder beryllium.
- Present design of annular MAPLE targets using either HEU or LEU oxide powder.
- HEU aluminum alloy in small diameter fuel/target rods as in SLOWPOKE 1 and 2.
- LEU oxide ceramic pellets in small diameter fuel/target rods as in SLOWPOKE 2.

4. Conclusions and future options

4.1 AECL is confident that NRU could continue operating until 2021 and possibly longer. However to guarantee reliable operation up to and beyond that date, it would be prudent to order a new reactor vessel now.

4.2 Without a major facility like NRU at Chalk River, the best and brightest young scientists and engineers will go elsewhere. **NRU may be old, but it is certainly not obsolete. It is still among the top five high-flux, multi-purpose, research reactors in the world.**

4.3 **In the long term, low-power aqueous homogeneous reactors might well turn out to be the most economic and most reliable sources of Mo-99.** The 200 kW homogeneous reactor currently being developed by Babcock & Wilcox in the USA, may provide the proof within the next 5 to 10 years. Meanwhile Canada could get a head start by converting an existing SLOWPOKE reactor to homogeneous operation. **Three 20 kW SLOWPOKE reactors or two 30 kW Chinese MNSR reactors could supply all of Canada.**

4.4 **The two MAPLE reactors were intended to be the first reactors in the world dedicated to commercial isotope production.** When they were shut down permanently in 2008, because of a technical problem which could not be solved without major redesign, a valuable Canadian asset suddenly became a liability, with both the taxpayer and private industry suffering huge losses. Consequently, Natural Resources Canada provided limited funding for research and development specifically directed to non-fission sources of medical isotopes, such as linear accelerators and cyclotrons.

4.5 When the Power Coefficient of Reactivity problem became acute, the present proposal to convert the 10 MW MAPLE to a 3 MW MAPLE-SP by eliminating the driver fuel, was unique among the many solutions proposed at that time; unique because **MAPLE-SP is a major simplification of the MAPLE concept without requiring a major redesign of the structural hardware.** An exception is the chemical processing system for extracting Mo-99 from LEU oxide fuel/targets. It will require costly redesign and engineering development.

4.6 MAPLE-SP targets can use LEU, by increasing the thickness of the uranium oxide annulus in the present MAPLE targets, which were designed for HEU.

4.7 Obviously the reactor and chemical plant are only two components in the complex Mo-99 production and distribution chain, but they are essential. **MAPLE-SP may be cost-effective, or it may not be, but at least it warrants serious consideration.**

5. References

[1] Mohamed Zakzouk, “The 2009 – 2010 Medical Isotope Shortage: Cause, Effects and Future Considerations”, Library of Parliament Publication No. 2009-04-E, Ottawa (revised November 2010)

[2] R. L. Gagnon & H. Bonin, “Homogeneous SLOWPOKE Reactor for the Production of Radioisotopes: A Feasibility Study”, Proc. 29th Annual CAN/CNS Student Conference, Toronto, Ontario, 2005

[3] R. E. Kay, J. W. Hilborn, P. D. Stevens-Guille and R. E. Jervis, CRNL-568 (Rev 5), Chalk River Nuclear Laboratories, Chalk River (1975)

[4] W. L. Cheng, C. S. Lee, C. C. Chen, Y. M. Yang and G. Ting, “Study on the Separation of Molybdenum-99 and Recycling of Uranium to Water Boiler Reactor”, Appl. Radiat. Isot. Vol. 4, No. 4, pp. 315-324, 1989

[5] Idele Esterhuizen, “Necsa Expands Isotope Production Study To Include Multi-Purpose Reactor”, Engineering News Online, South Africa (March 9th, 2012)

[6] C. B. Mills, G. A. Jarvis, “Critical Mass Reduction”, LA-3551, Los Alamos (1967)

6. Acknowledgements

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