Producing Molybdenum-99 in CANDU Reactors

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Abstract

This paper discusses the recent problems with Canadian supply of molybdenum-99 for medical diagnostic scanning. It proposes an alternate method that exploits the on-power refueling capability of CANDU reactors to produce large amounts of Mo-99. An extraction and refining plant near the used fuel bay of a multi-reactor station could process one standard fuel bundle per day (after irradiation for 5 days). This method avoids using enriched uranium. The plant might cost less than 50 million dollars and be constructed within several years. The radioactive residue would be managed in conjunction with the existing methods of used fuel management.

1. Introduction

During normal operation, the multi-purpose NRU research reactor in Chalk River Laboratories was producing about 30 percent of the world's supply of molybdenum-99 (Mo-99), a very important radionuclide because it beta decays ($T_{1/2}$ = 66 h) to technetium-99m (Tc-99m). The latter is used in diagnostic imaging with single photon emission computed tomography (SPECT) technology. Tc-99m is employed in about 80% of the nuclear medicine diagnostic procedures in Canada. Excellent information on this subject appears in the Report of the Expert Review Panel on Medical Isotope Production [1] that was commissioned by Natural Resources Canada.

NRU began operating in 1957 and started producing Mo-99 in the early 1970s. This heavy water reactor (thermal neutron flux: ~ 3×10^{14} n/cm^2/s) fissions highly enriched uranium (HEU) 'targets' for about a week. The targets, an alloy of uranium and aluminium, are transferred to a nearby processing facility where Mo-99[†] is extracted and transported to MDS Nordion in Ottawa. Concerns arose about relying on a 50-year-old reactor for this essential service. To address this anxiety, a project was started in the mid-1990s to build two dedicated MAPLE-type reactors with a facility to extract the Mo-99. Licensing, technical and economic problems were encountered while implementing this Dedicated Isotope Facility (DIF). In May 2008, the Government of Canada accepted AECL's decision to terminate this project. Realizing that the reliability of Canadian supply of Mo-99 is again a concern, the author began advocating that a *back-up* method of supply be developed that would utilize one of the many nearby CANDU power reactors. This would avoid the 10-year duration to build a new reactor and the very large project and operating costs that would be associated with such an endeavour.

CANDU reactors use natural uranium. During normal operation, robotic fueling machines load fuel bundles into one or two fuel channels every day. Over the years, many have thought about making Mo-99 in these power reactors; however, they were deterred by the complexity that would be added to the already challenging task of operating the reactors safely and efficiently.

[†] Mo-99 is a fission product with a cumulative yield of 6.11 per 100 fissions.

Also, there was no economic incentive because Mo-99 production in the NRU reactor has been highly subsidized by the Government of Canada.* The decision to stop DIF construction created considerable Canadian and world anxiety about supply of Mo-99.

In early 2009, the author conceived the idea of putting MAPLE-type HEU targets (annular tubes) into a bundle with the same form, fit and function (same power rating) as a standard 37-element CANDU fuel bundle. It would involve the following steps: identify a willing CANDU operator, fabricate HEU target bundles, test the design, and transport irradiated target bundles to the existing processing facility that extracts Mo-99.

2. NRU Shutdown and the Expert Review Panel

In mid-May 2009, a heavy water leak from the NRU calandria was detected and the reactor was shut down to investigate and repair the leak. Considerable outrage arose in the Canadian and worldwide medical community at the interruption in the supply of Mo-99, as reported in many media articles and broadcasts. At the annual Canadian Nuclear Society conference, 2009 May 31 to June 3, the author discussed the possibility of a back-up supply with personnel from a CANDU station operator, AECL and the nuclear regulator. This idea was neither dismissed nor endorsed. More information was requested.

The Government of Canada felt strong social and political pressures to address the problem promptly, and took a variety of actions on supply [2]. In mid-June, it established the Expert Review Panel on Medical Isotope Production (the Panel) to provide advice on the most viable options for securing a predictable and reliable supply of Tc-99m in the medium to long term [3]. At the time the Panel was announced, May 28, 2009, a call for Expressions of Interest (EOI) was put out to public and private sector organizations for submissions (by July 31) on alternative production of Mo-99/Tc-99m. The Proponent's Guide [4] was issued in early July. Twenty-two EOIs were received and assessed against five criteria that were established by the Panel. The author submitted an EOI that proposed producing Mo-99 in CANDU reactors along the lines outlined above.

The Panel's report was issued to the government on November 30, on schedule. The Report [1] describes the Panel's mandate, activities and processes. It covers the background very well: the nuclear history, starting with the use of radionuclides in medicine and Canada's role in this development. It outlines clearly the present structure of the worldwide Mo-99 supply system, which has been based mainly on the output from five government-owned and funded multi-purpose research reactors that were put into operation in the period from 1957 to 1966. They are located in Canada, Europe and South Africa. Consequently, the costs paid by the refining, packaging and distributing companies do not reflect the real costs of Mo-99 production, and this sets a low price for the Mo-99 that is supplied to the world medical community. The people of Canada have been subsidizing one-third of the world's supply. The Report discussed the market trends and how the future directions might change, depending on the duration of the shortage.

As a result of the interruption in the Canadian supply, the price has increased and the distribution of Mo-99 has changed in response to market demand. There has been significant diversion to

* Production of cobalt-60 in CANDU reactors has been profitable for plant operators.

North America. The increased cost and reduced availability of the radioisotope has challenged the world medical community, and it has adapted. In many cases, diagnostic examinations using Mo-99 have been cut back (deferred or cancelled). Different arrangements have been made, including performing essential diagnostic examinations using alternate techniques, some of which are identified in the Report. The interruption has had adverse health consequences.

The Panel assessed the options for Mo-99 production, identifying the two classes of technology, reactor (fission option) and accelerator (photo-fission, Mo-100 transmutation and direct Tc-99m using a cyclotron). The comparison looked at cost, timeline (to first production) and capacity (fraction of Canadian demand). The other factors addressed were sustainability and security, technical feasibility, business implementation, timeliness, regulatory issues and benefits to Canadians. The Panel reviewed all of the EOI proposals, but did not discuss each specifically. The report pointed out that options that depend on HEU could be viable only in the short to medium term.

The Report recommended replacement of the NRU reactor. The Panel believes that "a multi-purpose research reactor represents the best primary option to create a sustainable source of Mo-99, recognizing that the reactor's other missions would also play a role in justifying the costs." This option would cost between 500 million and a billion dollars. It would take about ten years to implement. (Priority for reliable Mo-99 production would compromise the other missions of this reactor for research.) The Panel also recommended support for an R&D program for cyclotron-based Tc-99m production. It advocated better use of Tc-99m supply through the use of newer medical imaging SPECT technologies and investment in positron emission tomography, to reduce the demand for Mo-99. Further discussion was provided on linear accelerator options and the DIF infrastructure at Chalk River Laboratories.

3. Workshop on Medical Radionuclide Production

In parallel with the Panel's activities, the Canadian Nuclear Society (CNS) organized a workshop on medical radionuclide production that was held in December in Ottawa [5]. All the participants in this event (that featured 15 presentations) gained a much better knowledge and appreciation of some of the methods and technologies being deployed in Canada and abroad to produce Mo-99 and carry out different types of diagnostic scans.

The U.S. demand and dilemma was explained in an excellent presentation from Sandia National Laboratories [6]. The radionuclide Tc-99m is used in about 13,000,000 medical diagnostic procedures each year in the U.S. Mo-99 consumption is 5000 to 7000 curies (6-day) per week.[*] This translates to 38,000 to 53,000 production curies per week, allowing one day for processing and shipping (specific activity > 5000 Ci/g of Mo-99 required). U.S. usage of Mo-99 has been increasing by 3 to 5 % per year. All major production uses HEU targets with the HEU supplied by the U.S. Concern about weapons proliferation is causing a change from HEU (93% U-235) to LEU (19% U-235) supply within 5 to 7 years (five times more target material to be irradiated).

A fission source of about 1.1 MW of continuous power in the targets would supply the nominal U.S. demand—about 2.2 MW (78 to 106 kCi per week) for the world demand [6].

[*] A 6-day curie is the amount of Mo-99 (τ = 95 h) activity that remains after 6 days (144 h) of decay.

4. Better Concept for Producing Mo-99 in CANDU Reactors

Since a CANDU fuel channel holds 12 bundles and outputs about 6.5 MW of power, four fuel bundles are a fission source of about 2.2 MW and could supply the world Mo-99 demand. The U-238 component in the fuel would not contribute significantly to the Mo-99 production in a short irradiation and would remain in the residue after Mo-99 extraction. More Pu-239 would be produced in natural uranium targets than in HEU/LEU targets; however, the total alpha-emitter concentration (considering the U-234 in HEU/LEU) would not be significantly higher for the short irradiation [6].

The following calculation was performed to check the production rate given in Reference 6. Since each fission reaction deposits about 175 MeV or 2.8×10^{-11} watt-s of energy (using the conversion factor: 1 MeV = 1.60×10^{-13} watt-s), four bundles undergo $(2.2 \times 10^{6}) \div (2.8 \times 10^{-11}) = 0.786 \times 10^{17}$ fissions/s. Multiplying this fission rate by the Mo-99 (cumulative) yield, 6.11 per 100 fissions, gives the production rate: 4.80×10^{15} nuclei/s.

The amount accumulated in 6 days (t = 144 h) is: $4.80 \times 10^{15} \times (1 - e^{-\lambda t})/\lambda$ nuclei/s, and the corresponding activity is: $4.80 \times 10^{15} \times (1 - e^{-\lambda t})$ Bq, where λ is the decay constant, 0.0105 h^{-1}. The Mo-99 activity in curies is: $4.80 \times 10^{15} \times 0.78 \div (3.7 \times 10^{10}) = 1.01 \times 10^{5}$ curies, which is comparable to the value 78 to 106 kCi given in Reference 6.

Because 1% of the Mo-99 produced decays away every hour, it is very important to locate the extraction and refining plant beside the reactor. The penalty is significant for off-site processing due to the time lost in target transport and the transport container expense. Batch processing, weekly removal followed by a week of processing, gives only 50% of the product that could be produced from daily extraction (of one fuel bundle) and daily processing. Time is also a factor in product quality; specific activity (curies/g) decreases with time after irradiation [6].

The radioactive residue from processing four bundles per week, or 210 bundles per year, could be managed along with the approximately 6000 used bundles that are removed every year from each power reactor. There would be no need to ship HEU/LEU and no concern about the unused U-235 accumulating at the site.* The energy from the targets would generate power. Producing Mo-99 in a multi-reactor station would avoid supply interruptions due to maintenance shutdown.

If construction of a Mo-99 processing plant at a CANDU station is considered, who could build it; how much would it cost and how long would it take to complete?

5. On-Site Processing Plant

An excellent presentation on this subject was delivered by INVAP of Argentina [7]. This company recently completed the OPAL Reactor Project (Figure 1). It included the Radioisotope Production Facility (Figure 2). The facility produces many important radionuclides, such as Mo-99, which is extracted from LEU uranium-aluminium alloy targets. The project was completed on schedule. The budget amount ($200 million) suggests that the cost of just the processing

* A one-week irradiation in a CANDU uses only ~ 10% of the HEU (much less in LWR), so the waste is still HEU.

plant at a CANDU station would be a small fraction of this amount. The design of the plant would be similar to the OPAL one, even though the targets would be different, uranium-oxide pellets in a zirconium alloy cladding. The process to extract Mo-99 from such targets is well known, so the expected construction time for a repeat plant would be several years.

6. Conclusion

Producing very large quantities of Mo-99 in CANDU reactors would be feasible and relatively inexpensive. The main requirement—the construction of a plant beside the used fuel bay of a multi-reactor station that would process one normal fuel bundle per day after five days of irradiation. The Mo-99 output would be very reliable and would be sold to existing distributors.

7. References

[1] The Expert Review Panel on Medical Isotope Production letter to Minister of Natural Resources Canada, "Report of the Expert Review Panel on Medical Isotope Production," November 30, 2009. Available at: http://nrcan.gc.ca/eneene/sources/uranuc/pdf/panrep-rapexp-eng.pdf

[2] Medical Isotopes, Government of Canada's Action on our Supply. Available at: http://nrcan.gc.ca/eneene/sources/uranuc/mediso-eng.php

[3] Natural Resources Canada news release, "Government of Canada Announces Expert Review Panel for Long-Term Isotope Supply Solutions," 2009/51, May 28, 2009. Available at: http://www.nrcan-rncan.gc.ca/media/newcom/2009/200951-eng.php

[4] "Expert Review Panel on Medical Isotope Production, Call for Expressions of Interest, Proponent's Guide," July 2009. Available at: http://nrcan.gc.ca/eneene/sources/uranuc/isotopes-guide-eng.php

[5] Canadian Nuclear Society, Workshop on Medical-Radionuclide Production Methods, Ottawa, December 11-12, 2009. Available at: http://www.cns-snc.ca/events/cns-workshop-production-medical-radionuclides/

[6] R. L. Coats, "Requirements for a Commercial-Scale Domestic Mo-99 Supply." Reference [5].

[7] M. Salvatore, "An INVAP Perspective on the Production of Medical Radioisotopes: Past & Present." Reference [5].

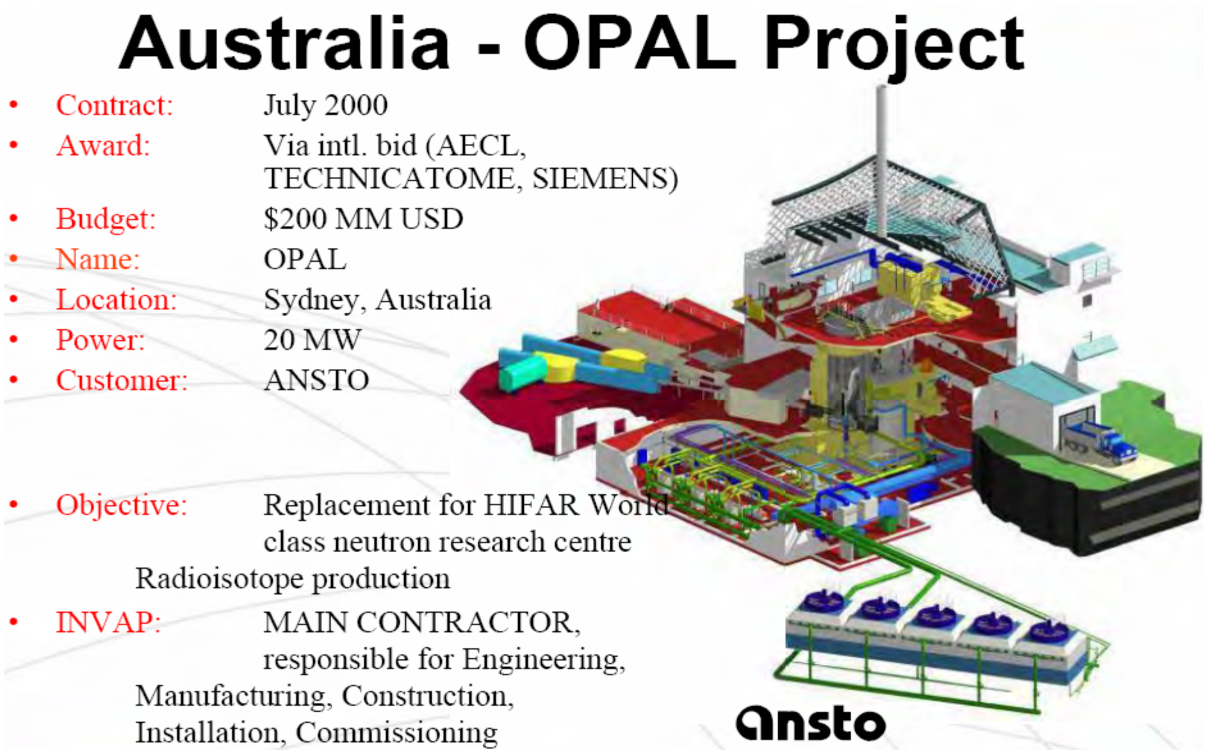

Figure 1. OPAL Project

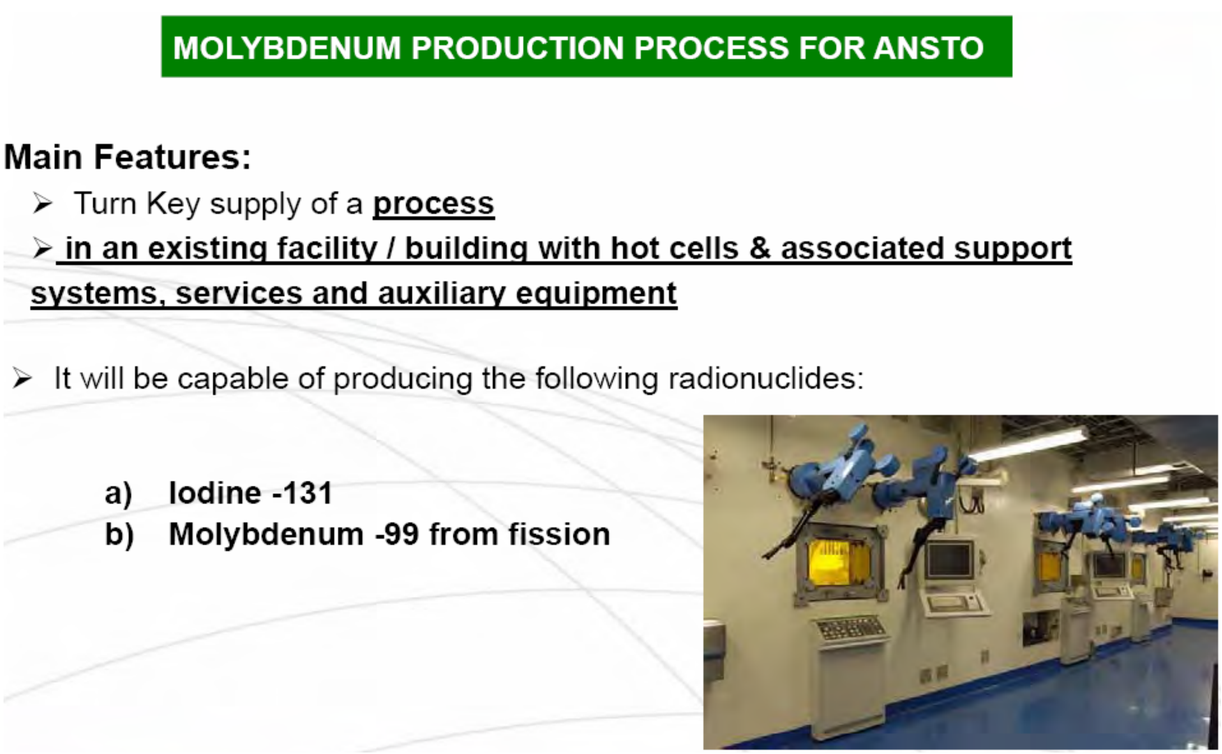

Figure 2. INVAP Mo-99 Production Facility at the OPAL Reactor