THE LEU TARGET DEVELOPMENT AND CONVERSION PROGRAM FOR THE MAPLE REACTORS AND NEW PROCESSING FACILITY

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ABSTRACT

Historically, the production of molybdenum-99 in the NRU research reactors at Chalk River, Canada has been extracted from reactor targets employing highly enriched uranium (HEU). A reliable supply of HEU metal from the United States used in the manufacture of targets for the NRU research reactor has been a key factor to enable MDS Nordion to develop a secure supply of medical isotopes for the international nuclear medicine community. The molybdenum extraction process from HEU targets provides predictable, consistent yields for our high-volume molybdenum production process. Each link of the isotope supply chain, from isotope production to ultimate use by the physician, has been established using this proven and established method of HEU target irradiation and processing to extract molybdenum-99.

To ensure a continued reliable and timely supply of medical isotopes, MDS Nordion is completing the construction of two MAPLE reactors and a New Processing Facility. The design of the MAPLE facilities was based on an established process developed by Atomic Energy of Canada Ltd. (AECL) – extraction of isotopes from HEU target material. However, in concert with the global trend to utilize low enriched uranium (LEU) in research reactors, MDS Nordion has launched a three phase *LEU Target Development and Conversion Program* for the MAPLE facilities. Phase 1, the Initial Feasibility Study, which identified the technical issues to convert the MAPLE reactor targets from HEU to LEU for large scale commercial production was reported on at the RERTR- 2000 conference.

The second phase of the *LEU Target Development and Conversion Program* was developed with extensive consultation and involvement of experts knowledgeable in target development, process system design, enriched uranium conversion chemistry and commercial scale reactor operations and molybdenum production. This paper will provide an overview of the Phase 2 Conversion Development Program, report on progress to date, and further discuss the challenges in converting the MAPLE facilities to molybdenum production from LEU targets.

The second phase will be completed by the end of calendar year 2003 and would lead into the third phase, the implementation of the *LEU Target Development and Conversion Program, w*hich, if technically and economically feasible, is expected it would be completed in 2007.

1. Introduction

It is important to step back and consider the context in which MDS Nordion's MAPLE facility will operate. MDS Nordion is the world's leading supplier of medical isotopes. The company is in the business of supplying isotopes used to conduct some 34,000 nuclear medicine procedures every day around the world, such as determining the severity of heart disease, the spread of cancer and diagnosing brain disorders.

There are over 100 medical applications for radioisotopes and some 80% of nuclear medicine procedures rely on just one isotope, molybdenum-99. Moreover, some of these procedures are performed using medical isotopes as soon as 41 hours after leaving the reactor. This is a real just-in-time business. As the radioisotope decays, MDS Nordion must get the product to the customer as quickly as possible. This is also a global endeavor. For example, some 5000 hospitals in North America depend on this supply each week; in Germany, its 850 hospitals; in Japan, its over 1000 hospitals and in Argentina, over 250 hospitals rely on our supply.

MDS Nordion's medical isotope business is also providing an exciting new platform in radioimmunotherapy. For example, novel ways to use a radioisotope to treat disease, such as for non-Hodgkin's lymphoma, a blood-borne cancer, are being developed. This exciting platform will expand the horizon for applications of medical isotopes.

At the end of the day, the MAPLE story, its planning, construction and the development work relating to LEU conversion, is about securing the supply of medical isotopes required by the international nuclear medicine community, and, ultimately, the thousands of patients who expect to receive their daily medical procedures.

2. The MAPLE Facilities – Securing the World Supply of Medical Isotopes

Why build the MAPLE facilities?

There are compelling reasons to build the MAPLE facilities. MDS Nordion supplies the majority of the world's isotopes. Notably today molybdenum-99 is the most extensively used isotope. However, new medical techniques are providing opportunities for iodine-131, and the utilization of iodine-125 and xenon-133 are growing. The NRU reactor owned by AECL at Chalk River has been operating since 1957 and producing molybdenum since the early 1970's. Today NRU also supplies other medical isotopes, including cobalt-60.

Molybdenum-99 supply in the pre-1980 era was supported by four capable suppliers: Cintichem and GE in the United States, IRE in Belgium, and MDS Nordion in Canada. MDS Nordion obtained our medical isotopes in the NRU and NRX reactors, owned by AECL. Some of the reactors used for isotope production have encountered insurmountable circumstances that resulted in their permanent shutdown. The GE reactor was found to be located on a seismic fault and it was shutdown in 1980; the Cintichem reactor experienced a contamination incident that resulted in its permanent shutdown in 1990; the NRX reactor in Canada was shut down in March 31, 1993 due to safety concerns. The global nuclear medicine community had serious concerns about supply reliability, which was created by the shutdown of these reactors, as there was no viable backup supply of medical isotopes.

A number of initiatives to broaden molybdenum-99 supply were taken. Mallinckrodt established a contract with HFR at Petten with MDS Nordion's help; IRE increased its production capacity to provide a larger volume back up; NECSA in South Africa began to produce molybdenum-99 in 1993. None-the-less, the concern of a supply shortage continued. If NRU were to shutdown, the nuclear medicine community would not be able to obtain the volume of molybdenum-99 that it required from the aggregate of other producers.

In response to concerns the nuclear medicine community had about the long term, secure supply of molybdenum-99, in 1996 MDS Nordion and AECL announced an agreement that will ensure reliable and economic availability of radioisotopes to hospitals and clinics worldwide. The agreement provided for construction of two MAPLE reactors and a high volume, commercial, first stage processing facility at AECL's Chalk River Laboratories. MDS Nordion will own the reactors and processing facility and be responsible for managing the business and developing the isotope production planning activities. AECL have been contracted to design, build, and operate the facilities on behalf of MDS Nordion. The MAPLE reactors will be the only reactors in the world totally dedicated to the commercial production of medical radioisotopes. The significant investment being made by MDS Nordion at the AECL site at Chalk River, will capitalize on the extensive infrastructure, expertise and experience of AECL and
MDS Nordion for the reliable, continuous supply of isotopes.

What are the MAPLE Facilities?

The MAPLE Facilities consist of two reactors and a processing facility to extract isotopes and manage the process waste. Figure I shows the MAPLE reactors and the New Processing Facility (NPF) building (these are the buildings with light coloured siding). The photograph also shows the NRU reactor in the background and the NRX reactor which was shutdown in 1992, to the right in the photograph.

Figure I: MAPLE reactors and New Processing Facility

The MAPLE reactors are 10 MW, open pool, light water reactors, designed for the sole purpose of producing medical isotopes. Figure II shows a cross section of the MAPLE reactors. The reactor has a compact core, about the size of a 90 litre (20 gallon) drum, and is surrounded by heavy water reflector tank. The reactor assembly consists of five major components: the inlet plenum; the grid plate; the core structure consisting of vertical flow tubes containing low enriched uranium (LEU) driver fuel bundles and high enriched uranium (HEU) target assemblies for medical isotope production; the heavy water reflector tank; and the chimney. The light water primary coolant enters the inlet plenum, flows upward through the grid plate, the flow tubes and fuel and target assemblies, and is directed back to the suction of the primary cooling pump via the outlet arms of the chimney.

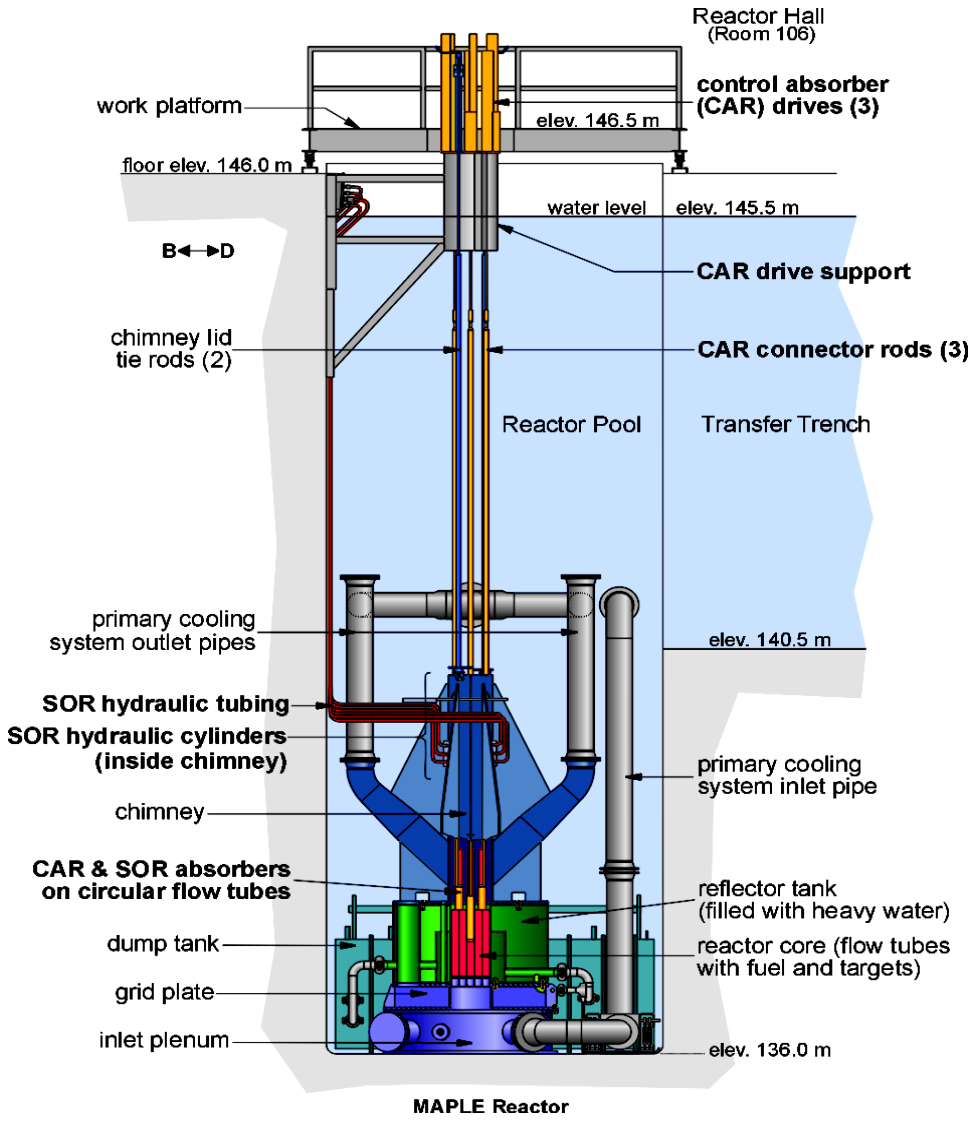

Figure II: MAPLE Reactors (SOR = Shut-off Rod, CAR = Control Absorber Rod

The MAPLE reactors and the isotope processing facility are licensed to irradiate and process HEU targets to produce the following medical isotopes as fission products of uranium-235: molybdenum-99, iodine-131 and xenon-133.

Figure III shows the MAPLE reactor core during low-power commissioning. The core is composed of 13 hexagonal and six circular flow tubes. Four of the 13 hexagonal flow tubes are used for irradiating HEU targets; the remaining nine contain 36 element LEU driver fuel assemblies. The six circular flow tubes contain 18 element driver fuel assemblies. A heavy water reflector surrounds the core and contains irradiation sites for the production of iodine-125.

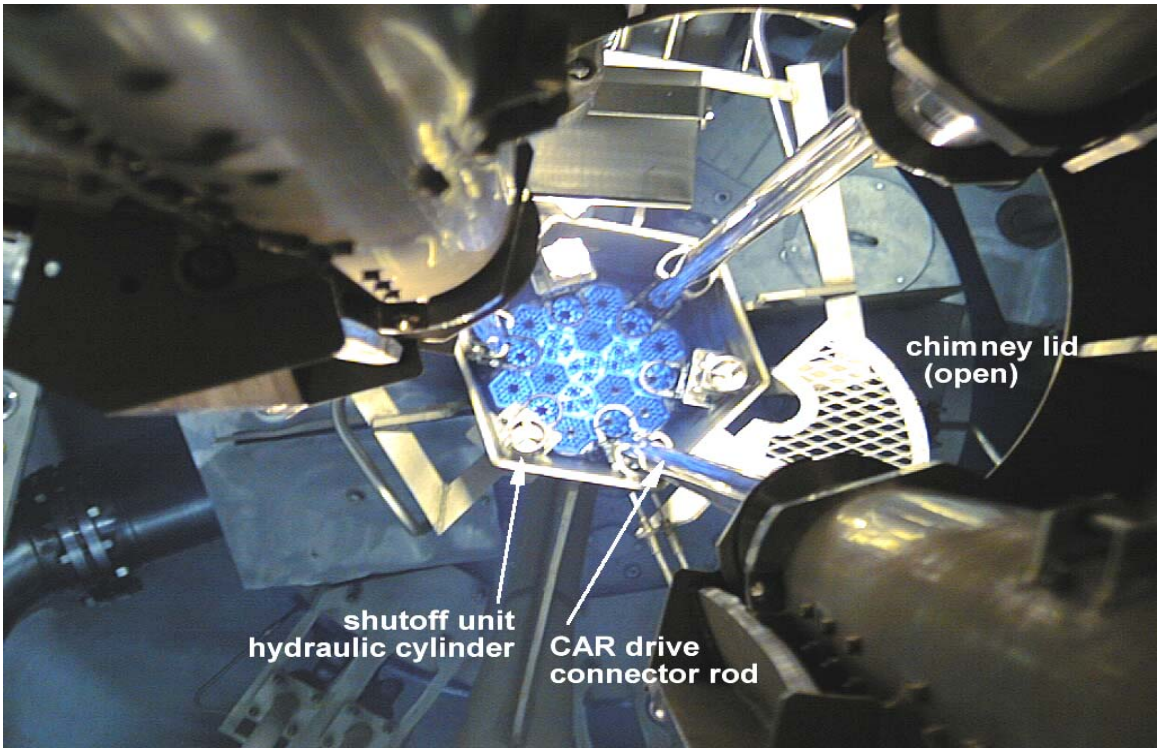

Figure III: MAPLE Reactor Core (CAR = Control Absorber Rod)

Two reactors were built to ensure a secure and continuous supply of medical isotopes. Isotopes will be produced in one reactor while the other is being maintained and on stand-by for its next production run. The operating cycle of each reactor will have a minimum operating period of up to four weeks depending on isotope demand. HEU targets irradiated in the MAPLE reactors will be transferred in shielded containers to the processing facility for isotope extraction. The radioactive waste from the extraction process will be solidified within the processing facility and transferred to the waste management area on the Chalk River site for storage in concrete canisters. Figure IV shows canisters used to store the waste. One canister is sufficient for storing waste from about 3 years isotope production with HEU targets.

Figure IV: Canisters for storing waste from isotope production.
Canisters in the background are used for storing spent reactor fuel.

The New Processing Facility will extract the radioisotopes produced in the HEU feedstock target material, process the residuals, and transfer the product to containers for shipment to MDS Nordion's Ottawa facility. There, the isotopes are further processed, packaged, and distributed to MDS Nordion's nuclear medicine customers around the world. The timeline from start of target processing to product delivery to the hospital can be as little as 41 hours.

Overall, completion of the MAPLE project was planned to be about fifty months in duration. These new, one-of-a-kind facilities had several challenges to meet during the execution of the project. Advanced technology, a new licensing environment and a compressed schedule created challenges in licensing, design, construction and commissioning. Construction of the facilities had to take place on a crowded site. Existing buildings had to be removed and the ZEEP reactor had to be decommissioned and dismantled**.** So far several key milestones have been achieved.

• Project Start	September	1996
• Environmental Approval	April	1997
• Construction Approval	December	1997
• MAPLE 1 Reactor		
- Operating License	August	1999
- First Sustained Nuclear Reaction	February	2000
• New Processing Facility		
- Operating License	August	1999
• MAPLE 2 Reactor		
- Operating License	June	2000

Figure V: MAPLE Project Key Milestones

An environmental assessment for the facilities was completed in April 1997 and construction approvals for these facilities were granted in December 1997. MAPLE 1 achieved its first sustained nuclear reaction on February 19, 2000. This was a significant milestone in the project. The MAPLE 1 reactor had achieved all of its performance objectives at the 2 kW level and power was increased to the 500 kW level. Also, the operating license for MAPLE 2 was received in June 2000. Since July 2000, there has been a pause in commissioning to address licensing issues related to the shut-off rod safety shutdown system.

Each MAPLE reactors has two independent and diverse safety systems.
(See Figures II and III.):

- Safety System 1 - has three hydraulically actuated shut-off rods.
- Safety System 2 - has three electromagnet actuated control absorber rods and a hydraulically actuated reflector dump system.

Any two of the three shut-off rods that are dropped into the core will place the reactor in a stable sub-critical state. Similarly, any two of the three control absorber rods that are dropped into the core will place the reactor in a stable sub-critical state. The reflector dump system will also place the reactor in a stable sub-critical state. Extensive testing of the MAPLE reactors safety systems has been completed and, after extensive review by the Canadian Nuclear Safety Commission, AECL is preparing to resume nuclear commissioning of the facilities which are expected to be in operation in calendar year 2003

3. Converting the MAPLE Facilities to LEU Targets

Tremendous progress has already been made by AECL and MDS Nordion to convert medical isotope production to LEU based material. The MAPLE reactors were designed to operate with LEU fuel, thus achieving a significant technological advancement in conversion from HEU to LEU. This in itself has reduced the reliance on HEU and thus makes the MAPLE technology unique at this point in time. The leadership taken by AECL and MDS Nordion to use LEU fuel in the MAPLE reactors for commercial production of medical isotopes is a substantial accomplishment in the Reduced Enrichment for Research and Test Reactors (RERTR) program.

HEU target technology is an integral part of the reactor operating system. The technology for production of large quantities of molybdenum-99 from HEU targets is based on a reliable

process that has been proven for some thirty years in reactors operated by commercial isotope producers. Predictable, consistent yields of molybdenum from HEU targets are the foundation for a reliable supply of medical isotopes. Furthermore, all of the requisite licensing has been approved by national nuclear regulators and by health care regulators such as the U.S. Food and Drug Administration (FDA), and European national authorities. These links provide a secure chain of medical isotopes for the international nuclear medicine community. For this reason, the isotope production process has been designed using HEU targets clad in zirconium alloy.

Although HEU target technology is well established and consumes a relatively small quantity of material, proliferation concerns have caused significant international interest in reducing reactor reliance on HEU feedstock material. Today, the only known commercial sources of HEU are the United States and Russia. South Africa has their own supply of HEU used for molybdenum-99 production. Concerns with international safeguards and non-proliferation of HEU material have caused the U.S. to enact legislation in that will encourage reactor operators to convert to LEU and create increased difficulty for non-U.S. organizations to access HEU from the United States. Known as the Schumer Amendment, changes were made to the Atomic Energy Act in 1992 that imposed restrictions on exports of HEU fuel and targets from the United States, with certain qualifying statements related to the technical and economic viability of converting to LEU.

Since the unfortunate circumstances around the incidents of September 11, the international community has heightened interest in security measures to ensure nuclear materials are well protected. In fact, the international nuclear regulatory infrastructure has enabled specific measures to ensure security measures are in place. For example, the USNRC, on a recent export license, has placed a condition that "If, while within U.S. jurisdiction, the material to be exported under this license is not protected in transit utilizing the services and equipment of the Department of Energy (DOE) National Nuclear Security Administration (NNSA) Office of Transportation Safeguards (OTS) in accordance with the DOE/NNSA requirements and directives for the transport of such material, then the licensee must comply with applicable 10 CFR part 73 regulations on physical security measures including any NRC-required enhancements thereto and must obtain NRC's approval of the physical security plan for each shipment."

Furthermore, there is today increasing difficulty in the logistics of sourcing HEU material worldwide. Generally, HEU material is government controlled, lead-time to access material is long, and the reliability of supply is uncertain. Although Canada is not a proliferation risk, international co-operation to comply with current legislation has caused MDS Nordion to initiate a program to examine the feasibility of, and address the issues to converting those reactors and their associated processing facilities to operate and process LEU targets. A significant challenge will be to convert the MAPLE facilities from HEU to LEU targets while they are operating; to ensure there is no disruption in the supply of molybdenum-99 and other radioisotopes used annually in millions of essential medical procedures.

To convert the MAPLE facilities to LEU targets in a logical manner, MDS Nordion established a three-phase *LEU Target Development and Conversion Program*. They are:

- Initial Feasibility Study
- Conversion Development Program
- Conversion Implementation Program

At the completion of each of the first two stages, an assessment will be done of the technical and economic issues that will be involved in the implementation phase.

It is important to understand that the *LEU Target Development and Conversion Program* must be accomplished within certain conditions. The first condition is that there must be minimum change to the MAPLE reactor design and operation, as well as the downstream processing and waste management systems, all of which have been designed, licensed and built based on HEU target technology. A second condition is that isotope production capacity must be maintained to ensure the production capability exists to meet market demand. As a consequence of these two conditions, the same number of targets will be used. As a result of this, the mass of uranium in the LEU targets will be 4.7 times greater than in an HEU target. The processing facility must be able to handle the increased uranium mass from the LEU targets and achieve acceptable performance characteristics in the areas of uranium dissolution, molybdenum-99 recovery yields, waste solidification and waste storage. Any incremental operational burden placed on the isotope production and processing system must ensure that the rigorous equipment preventative maintenance program for these facilities is not compromised.

4. The Phase 1 Initial Feasibility Study

The Phase 1 Initial Feasibility Study was completed in 2000 and significant results were yielded from it. A conceptual design for a LEU target for the MAPLE reactors has been produced and it has been determined that operation of the MAPLE reactors with LEU targets is technically feasible. The Initial Feasibility Study also identified the key Canadian regulatory conditions that must be met to use LEU targets in those reactors. Before LEU targets may be used in the MAPLE reactors, the Canadian Nuclear Safety Commission (CNSC) must review and approve environmental assessments and safety analyses performed by AECL, including critical heat flux tests and irradiation tests. It is anticipated that a public licensing process could be carried out by the CNSC in connection with its consideration of whether the MAPLE reactors will be authorized to use LEU targets. Based on consultation with the CNSC, it is expected that completion of their licensing process could require a minimum of three years. In addition, the drug certification requirements of the FDA and the European national authorities must be satisfied for production of molybdenum-99 from a new target source material comprised of LEU.

The situation with the New Processing Facility is more challenging and complex. Assuming that LEU targets could be irradiated in the MAPLE reactors, 4.7 times more uranium than in HEU targets will be chemically processed in the NPF to extract a similar quantity of medical isotopes. This additional mass must also be calcined to solidify the waste in stable form for long-term storage. The isotope processing hot-cell system and equipment, as noted earlier, is custom designed and solely dedicated to the processing and extraction of molybdenum-99 from HEU

targets. Therefore a thorough assessment of the processing facility was required to explore the technical viability of converting to LEU.

MDS Nordion commissioned an assessment of the New Processing Facility with the following three key objectives:

- determine whether the equipment designed for the NPF can process LEU targets;
- determine the production capacity with LEU targets; and
- determine changes that should be implemented prior to the introduction of radioactivity
  into the NPF.

The Phase 1 Initial Feasibility Study indicated the following key areas of concern in the New Processing Facility:

Liquid Waste Storage Capacity

The NPF has been built with adequate tanks to store the waste to reduce decay heat prior to the start of calcination. To improve production capacity, two areas have been identified: reduction of decay time and improvement of the calcination process.

Waste Calcination

The process used is a continuous batch process with the calcination taking place in a storage can that will be sealed and disposed of as solid waste. The calcination process requires the liquid from the target dissolution and alumina column washes to be metered into a can so free water can be evaporated on-line in a controlled manner. The system must operate under stable conditions to maintain process control; this will retain the residual material in the storage can, minimize sputtering which could cause fouling of the calcining equipment and minimize entrainment of the off-gas. Once the free water has been evaporated, thermal energy is applied to the residual material and a solid, cinder-like mass is left in the storage can. The storage can is then seal-welded, loaded in a basket, and transported in a shielded container to a storage silo in the waste management area. Because of the sputtering observed after the removal of free water, the total amount of uranium in the can is the limiting factor. With greater amounts of uranium arising from processing LEU targets, there was danger of sputtering not being contained in the can and causing fouling of the calcining system. The Initial Feasibility Study confirmed that a significant number of additional waste cans would be required if the change to LEU was made and the calcination process performed in a controlled manner.

A key step in the conversion feasibility evaluation is for MDS Nordion to gain operational experience processing HEU through the calcination and waste handling system to determine any changes that could be made to manage the increased mass of uranium that will be in the LEU targets. In addition, an evaluation will be made to determine if the process could be carried out more quickly. Furthermore this facility was built with an expected useful life of 40 years. In the natural course of events it is expected that MDS Nordion and AECL will find ways to improve the process capacity of the facility to accommodate any additional process demands to meet market growth projections.

Solid Waste Storage

Because of the processing considerations mentioned earlier, the number of waste cans will increase. As there will be less uranium-235 per waste can, the arrangement of these cans in the concrete waste storage silos (Figure IV) may be different and it is expected that the number of silos required will be greater. This, in turn, could have an impact upon the storage capacity of the existing waste storage site. The licensing and environmental requirements to address this issue will need to be defined.

Overall, the Initial Feasibility Study demonstrated that a LEU target could be processed and a similar molybdenum-99 extraction efficiency could be obtained as when compared to an HEU target. However, although there would be a greater volume of liquid waste required to obtain a suitable extraction efficiency, the limiting factor in system capability was identified to be the speed at which waste from the LEU target could be calcined. It was determined that this could best be addressed by establishing a process development program aimed at improving the cycle time to process the LEU waste stream. Essentially the key issue is the capacity and capability of the calcination system. This assessment was discussed with and reviewed by Argonne National Laboratory (ANL) who are co-operating with MDS Nordion and AECL on the LEU conversion program.

5. Status of the Phase 2 Conversion Development Program

With completion of the Phase 1 Initial Feasibility Study, MDS Nordion has proceeded with Phase 2, the Conversion Development Program. The Conversion Development Program is examining ways to address the two main obstacles to LEU conversion. These are:

1. high volumes of solution to separate the molybdenum-99 from the LEU targets because of the greater amount of uranium in solution; and
2. approximately five times more uranium will be present in the wastes to be calcined.

The Phase 2 program essentially consists of a waste process development program which is examining the technical, regulatory and economic implications for managing the increased volume of waste arising from processing LEU targets in the NPF. The objective of the Conversion Development Program Phase is to identify and evaluate improvements to the calcining system capacity and capability to process the LEU targets. The waste processing development program will:

1. identify throughput and cycle time improvements to the calcining system and equipment;
2. identify possible process improvements in the NPF to reduce the waste arising from processing LEU targets; and
3. ensure adequate equipment operation and maintenance cycles for commercial production.

The Phase 2 Program was formally launched by MDS Nordion in January 2001, in partnership with ANL, the Société Générale pour les techniques Nouvelles (SGN) and AECL. The program is scheduled to be completed at the end of calendar year 2003 and is progressing on schedule. Extensive progress has been made in Phase 2 on experimental and development work, examining ways to improve throughput and capacity of the calciner and waste management systems. ANL have played a key role in examining various precipitates as a means to improve throughput. This work has been substantially supported by the other partners in Phase 2 Conversion Development

Program. The designers of the waste management system, SGN, have examined technologies to improve calciner throughput and processing timeline. The process system integrators, AECL, are providing their overall expertise to ensure the entire process can continue viably as a full scale, continuous commercial operation.

Technical evaluations, bench chemistry precipitation and calcinations studies have been completed. The Phase 2 Program is currently in the engineering development phase, with operational process improvements and regulatory milestones for implementing a conversion program yet to be addressed. The primary requirements to be determined are the Canadian nuclear regulations as defined in the *Nuclear Safety and Control Act* and the FDA regulations related to use of drug products.

6. Conclusion

MDS Nordion is fully committed to continue to provide a secure, reliable source of isotopes to the international nuclear medicine community. To comply with the spirit and intent of policy and legislation intended to reduce reliance on HEU material, we have already made a significant contribution to the RERTR Program by having the MAPLE reactors designed to operate using LEU fuel. Furthermore, we continue to make progress on our *LEU Target Development and Conversion Program.* A key outcome will be the evaluation of the technical and economic feasibility of conversion program options at the end of Phase 2. The option chosen to convert to LEU targets must be both technically and economically feasible, and must ensure the reliable supply of medical isotopes, particularly molybdenum-99.

In the end, wherever our LEU conversion process leads, MDS Nordion will not lose sight of the fact that there are millions of individual patients that depend on our supply of medical isotopes every year.