Developing the Sandia National Laboratories Transportation Infrastructure for Isotope Products and Wastes*

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The U.S. Department of Energy (DOE) is in the process of preparing an Environmental Impact Statement for the production of molybdenum-99 (⁹⁹Mo) at one of four alternative sites including Sandia National Laboratories (SNL), Oak Ridge National Laboratory, Idaho National Engineering Laboratory, and Los Alamos National Laboratory. Although SNL has been indicated as the preferred site, the final selection of a site awaits the completion of the National Environmental Policy Act process.

BACKGROUND

Certain radioactive isotopes for North American and especially the United States' needs are enormously important to the medical community and their numerous patients. The most important medical isotope is ⁹⁹Mo, which is currently manufactured by Nordion International Inc. in a single, aging reactor operated by Atomic Energy of Canada, Ltd. The reactor's useful life is expected to end at the turn of the century. Production loss because of reactor shutdown possibilities prompted the U.S. Congress to direct the DOE to provide for a U.S. backup source for this crucial isotope. The SNL Annular Core Research Reactor (ACRR) was evaluated as a site to provide ⁹⁹Mo initially and other isotopes that can be economically extracted from the process. Medical isotope production at SNL is a new venture in manufacturing. Should SNL be selected and the project reach the manufacturing stage, SNL would expect to service up to 30% of the U.S. market under normal circumstances as a backup to the Canadian supply with the capability to service 100% should the need arise. The demand for ⁹⁹Mo increases each year; hence, the proposed action accommodates growth in demand to meet this increase. The proposed project would guarantee the supply of medical isotopes would continue if either the irradiation or processing activities in Canada were interrupted.

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Radioisotope manufacturing by both the DOE and U.S. companies is not new. The DOE has produced radioisotopes for a spectrum of users both public and private for several decades. Cintechem, Inc., manufactured ⁹⁹Mo and several other isotopes for a substantial U.S. market fraction as recently as 1989. The process used by Cintechem was approved by the Food and Drug Administration (FDA) and is described in a Drug Master File (DMF). DOE obtained the rights to the patented process and the DMF in 1991 and is proposing to use this process to save the time and expense of developing a new DMF. The DOE purchase included not only the process but also associated equipment including packages for product, wastes, and spent fuel.

The proposed plan comprises fabrication of unirradiated targets to Cintechem specifications, followed by irradiation in the SNL ACRR. The irradiated targets contained in a transfer cask would be moved to the adjacent Hot Cell Facility (HCF) for processing where the radioisotopes of interest would be separated from the fission product inventory. The short half-life product, ⁹⁹Mo in NaOH, would then be air transported to radiopharmaceutical (RP) manufacturers. Waste generated during the fabrication process would be temporarily stored on-site and then disposed of at proper waste facilities.

NUCLEAR MEDICINE

Nuclear medicine (Saha 1992) is that medical community branch that applies the knowledge and materials of radioactive phenomena to the practice of both diagnosis and therapy. The seminal "tool" of nuclear medicine is the RP which is typically a compound consisting of a radioisotope and some specific medical agent such as a protein. However, radioactive elements themselves may be used as RPs, for instance using xenon-133 (¹³³Xe) in lung ventilation studies. By far, most RPs are used for diagnosis with some estimates as high as 95% while the remainder are used for therapeutic treatment.

In designing an RP, a pharmaceutical is chosen on the basis of its preferential location in a certain organ or its activity in the physiologic function of the organ. A suitable radionuclide is tagged onto the selected pharmaceutical so that after administration of the RP, radiation emitted from it may be detected and its location identified. The importance and uniqueness of RP procedures stems from the fact that resultant scans reveal organ functioning.

The isotope technetium-99 (^{99m}Tc), which is the decay product of ⁹⁹Mo, is a medically attractive element in its metastable form. The metastable state of a nucleus is at a higher energy potential than the ground state (Chase and Rabinowitz 1968) and it is this phenomenon that produces gamma rays upon transition that is so useful in medical detection apparatus. The radiation decay characteristic peak at 140 keV provides an energy which can pass out of the body and is compatible with detectors because this is within their window of efficiency of detection. The half-life of ^{99m}Tc is about 6 hours making it a very useful, yet perishable, substance.

Medical dispensation of ^{99m}Tc is accomplished with the ⁹⁹Mo-^{99m}Tc generator (Saha 1992). Within the generator, ⁹⁹Mo is adsorbed on alumina after which the ^{99m}Tc will

grow by the decay of the ⁹⁹Mo until maximum activity is reached after approximately four half-lives of ^{99m}Tc. At equilibrium and thereafter, the ^{99m}Tc radioactivity follows the half-life of ⁹⁹Mo. The ^{99m}Tc radionuclide is eluted as sodium pertechnetate (Na ^{99m}TcO₄) with a 0.9% NaCl solution. After elution the ^{99m}Tc radioactivity grows again and elution may be carried out even before the equilibrium is achieved. It is this substance that medical personnel prepare and use for numerous and varied purposes.

The perishability of these products, both the bulk materials and the derivative medicines, demand rapid handling and packaging, and a transportation system that is highly efficient and timely.

REACTOR, TARGETS, AND TRANSFER

There are several reasons why DOE considers SNL's ACRR and associated facilities to be a promising site for this isotope production program (DOE 1995). The ACRR is (1) a modern facility; (2) presently in an operational state with characteristics that are compatible with radioisotope production; (3) capable of being dedicated to continuous isotope production, which is necessary to meet the demands for short-lived medical-use isotopes; (4) collocated with the HCF and both can be modified with relative ease; and (5) in close proximity to excellent air transportation facilities (Albuquerque International Sunport) for radioisotope shipments.

The ACRR facility, consisting of the reactor and all support systems required for its operation, is located in Technical Area V (TA-V) at SNL. The ACRR became operational in 1978 and was originally designed with characteristics suitable to support weapons programs (Reuscher et al. 1982). The ACRR's capability to have large-volume, thermal-flux traps makes it a viable resource for producing radioisotopes, especially for those isotopes produced in the fission process.

A sketch of the ACRR is shown in Figure 1. The reactor core is installed in a large open tank filled with about 10 meters (m) (33 feet [ft]) of water to provide both core cooling and radiation shielding. The core is cooled by natural convection in an open water pool; the water pool is cooled by an external heat exchanger. The current ACRR configuration consists of an annular array of UO_2 -BeO-fueled elements having an active fuel height of 52 centimeters (cm) (20.5 inches [in.]). The dry, steel-lined, control cavity would be removed from the center of the core to provide a flooded region for target irradiation. Configurations of up to 19 or 37 targets at a time are planned. The targets would essentially fuel the reactor, therefore, only 180 or 130 conventional fuel assemblies, respectively, would be required.

The ACRR currently operates in either a pulsed or steady-state mode. The current steady-state power limit is 2 megawatts (MW). This limit is due to the combined heat rejection limitations of the heat exchanger/cooling tower system. With installation of additional heat exchanger/cooling tower heat rejection capacity would allow the reactor to run at 4 MW. For isotope production the ACRR would be operated in the steady-state mode and operations would be at or below 4 MW.

Because the ACRR is a pool reactor, targets and fuel elements would be readily accessible for removal. Targets consist of stainless-steel tubes approximately 45 cm (18 in.) long and 3.18 cm (1.25 in.) in diameter, as shown in Figure 2, containing highly enriched ²³⁵U that is a layer of uranium oxide (U_3O_8) approximately 50 microns (5.0 x 10^{-5} meters) electroplated onto the inside surface of the tube. They will be irradiated to provide a range of fission products including isotopes of molybdenum, xenon, and iodine. Targets irradiated for several days would be removed from the core and transferred via pass-through ports to a rack in the Gamma Irradiation Facility (GIF) pool (see Figure 1). A transfer cask would be lowered into the GIF pool and the irradiated target(s) would be loaded into the cask and transferred to the HCF using a manned transport vehicle.



Figure 1. ACRR Reactor Tank and Pool Cooling



The HCF will be reconfigured to streamline the process of irradiated target processing. One of the proposed HCF modifications to allow for sufficient extraction operations is the addition of new Steel Confinement Boxes (SCBs) that would result in safer, more reliable, and more versatile operations. The new SCBs would be designed to provide complete process control, including waste minimization and management. The units would be designed to collect byproducts from the radioisotope extraction, process the byproducts, and package them into waste containers. The design of the SCBs would be modular to allow easy replacement of components. Another important addition to the HCF is the Quality Control Laboratory that is required by the approved FDA procedure.

The irradiated targets containing almost 20,000 curies (Ci) of fission products would be processed within the SCBs. The isotopes of interest would be extracted from the fission product inventory by chemical dissolution and precipitation procedures in which: the noble gases and iodine would be condensed from the target fill gas; fission products would be dissolved from the inside of the target; chemicals would be added to maintain specific fission products in solution; molybdenum would be precipitated, filtered, and cleansed; and the precipitated molybdenum would be redissolved for shipment to pharmaceutical companies.

Although ⁹⁹Mo is the initial product of interest, ¹³¹I and ¹³³Xe may be directly extracted from the processing line as additional medical-valued products. The isotope ¹²⁵I may also be processed from ¹²⁴Xe, a nonradioactive isotope of xenon; however, this process requires additional apparatus and will only be explored after sufficient success is achieved in ⁹⁹Mo processing.

Each target would yield up to 800 Ci of ⁹⁹Mo, 200 Ci of ¹³¹I, and 600 Ci of ¹³³Xe one day after discharge from the reactor. The isotopes would be further purified to meet FDA DMF standards. The isotopes would then be properly packaged and shipped in shielded casks by air freight to RP companies. Approximately 20 to 25 targets per week can meet 100% demand with proportionately less targets for the anticipated level of 30% U.S. demand. It should be noted that the ACRR will have the capability to irradiate up to 37 targets continuously, but this level would be used only under extraordinary conditions of national need and would place a greater burden on the processing facility.

Approximately one-half liter (about 1 pt) of neutralized process liquid (waste) would be generated per target. This liquid would be solidified, allowed to decay, and then shipped to an approved low-level waste disposal facility. Although shipments to the disposal site could be made as soon as 6 months after generation, sufficient storage exists within the HCF to allow up to 2 years storage of waste generated as a result of meeting 100% of U.S. demand.

PACKAGING AND TRANSPORTATION

Table 1 shows the general characteristics of the three types of packages that will be used in the Isotope Production Project (IPP). The DOT- and NRC-certified Type B (NRC 1993a) package designated for use in transporting ⁹⁹Mo and ¹³¹I is the CI-20WC-2 or -2A model (NRC 1993b). The primary difference between the two models is the size and the amount of shielding. Both CI-20WC (Figure 3) models are described as follows: steel encased, wooden outer protective jackets with a depleted uranium shielded cask, and inner steel containment vessel. The protective jackets are contained within an 18-gauge steel drum. The inner containment vessel is a 2.73-in.-OD x 5.56-in.-long (7-cm-OD x 14.26-cm-long), 416 stainless-steel, gasketed and threaded container. The contents type and form of material for which the packages are certified are: ⁹⁹Mo/⁹⁹Tc in normal form as solids or liquids with a maximum quantity of material per package of 1,000 Ci, and ¹³¹I in normal form, or liquids with a maximum quantity of material per package of 200 Ci. Although the packages may contain the same maximum quantity of material, the Transportation Index (TI) will differ for each with a greater TI for the smaller package given the same amount of isotopic substances.

The package designated for transportation of product waste is the NRC-certified B-3 Type B package (NRC 1990). The packaging consists of a 6-in. (15.38 cm) lead shielded steel weldment in the shape of a right hollow cylinder with a bottom and a recessed, plugtype gasketed and bolted lid. Packaging features include lifting and tie-down devices and a drain to the central cavity. The maximum weight of the loaded package is 30,000 lb (13,636 kg).

Characteristics	Product		Waste	Spent Fuel
	CI-20WC-2	CI-20WC-2A	B-3	BMI-1
Cavity Diameter Cavity Length Shielding	3.1 in. (7.95 cm) 6.0 in. (15.38 cm) 2.2 in. (5.64 cm) DU + .35 in. (.9 cm) Steel	3.1 in. (7.95 cm) 6.0 in. (15.38 cm) 1.95 in. (5 cm) DU + .95 in. (2.43 cm) Steel	26.5 in. (67.95 cm) 43.25 in. (110.9 cm) 6 in. (15.38 cm) Pb + 1.25 in. (3.2 cm)	15.5 in. (39.74 cm) 54.0 in. (138.46 cm) 8 in. (20.51 cm) Pb + .87 in. (2.23 cm)
Weight, Empty	440.5 lb (200.2 kg)	314 lb (143 kg)	Steel 21,000 lb (9,545 kg)	Steel 21,860 lb (9,936 kg)

Table 1. Type B Package Characteristics



Figure 3. CI-20WC Transport Package

Spent nuclear fuel shipments are not expected to occur for several years and such fuel will be stored until DOE designates a repository. When shipments are to be made the BMI-1 will likely be used. The package assembly consists of five major components: the stainless-steel enclosed lead shielded cask, the stainless-steel encased lead shielded cover and gasket, the radioactive material, internal canister or basket, and a custom matching skid on which the cask rests vertically.

Packaging for ¹³³Xe will likely be a Cintechem-designed Type A gas cylinder. The package for ¹²⁵I has not been determined.

PRODUCT AND WASTE SHIPMENT

Isotope ⁹⁹Mo decays at the rate of about 1% per hour. Consequently, shipment of the product must be expedited to prevent needless decay of the product. Nordion, which is the only North American company presently shipping bulk ⁹⁹Mo, uses a combination of commercial and chartered air flights.

We expect to ship up to 800 Ci of ⁹⁹Mo per processed target and about six to seven packages per week at the nominal 30% production level. The ⁹⁹Mo product will be pharmaceutical quality and an FDA DMF will be existent for its production. At the present time, ¹³¹I and ¹³³Xe have DMFs and their shipment initially would likely occur for sampling purposes. The ⁹⁹Mo is expected to be shipped initially on a daily basis to one of three different locations: St. Louis, Chicago, and Boston. Air freight express class of shipments are envisioned. If a stop is required, the shortest time of routing from Albuquerque to the customer city will be preferable. Product movement from the TA-V reactor area to the airport transfer point using Kirtland Air Force Base and Albuquerque International Sunport access roads is the preferred route, avoiding public roads. Quality assurance (QA) may occur during the time the product is in shipment.

The primary waste disposal site designated for production and laboratory wastes is the Nevada Test Site facilities north of Las Vegas, Nevada. The site is compatible for these classes of waste and the site is operational. Two alternative waste disposal facilities are located at the Hanford Site near Richland, Washington. The 200-West Waste Generation Facility is quite extensive and has several current burial sites as well as some older burial grounds that are monitored. A second, alternate site is operated by U.S. Ecology and it is a commercial facility.

Initially, it was thought that two containers could be shipped on one truck. Further research has determined that an overweight truckload may result and could not be authorized; therefore, one truck would carry one B-3 package per shipment. The shipments would go directly from the ACRR building to either the primary site, or to either alternative site along the most direct route selected by the motor transport company. Approximately 85 shipments per year will be required. The dose criteria is the same as stated above for the product transport package as this package is Type B and must meet the same listed constraints.

All packaging used in the IPP will adhere to the requirements of the DOT as specified in the applicable parts of 49 CFR (DOT 1994).

SUMMARY

The growing importance and versatility of nuclear medicine are but a few of the reasons for an expanding market both in the United States and worldwide. As populations continue to grow, so will the need for nuclear medicine. New combinations of radionuclides and medical substances are increasing the spectrum of RPs available to the medical community. Concurrently, as the technology is expanding the number of countries acquiring such technologies is expected to increase, perhaps dramatically. A sharply increased demand for RPs could result.

Future considerations of loading, handling, and movement of both product and waste containers should address the need for modified or new containers and those that are suitable for automation. Many benefits can accrue by minimizing the human element in operations, notably as low as reasonably achievable concerns and the possibility of human error. In addition, other enhancements such as real-time cask identification survey, and real-time radiation survey could be safely and quickly carried out. Planning for these capabilities should be accomplished simultaneously with other automation efforts that could be applied to hot-cell processing.

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