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RECLAMATION OF GREATER THAN CLASS C SEALED SOURCES AT THE LOS ALAMOS NATIONAL LABORATORY

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ABSTRACT

One of the important overriding themes of the Los Alamos National Laboratory as a world-class scientific institution is to utilize its expertise in enhancing the long-term welfare of society by minimizing negative side effects of nuclear technology over the past five decades. The Los Alamos National Laboratory is therefore committed to the use of its technical competencies and nuclear facilities, developed through programs in the areas of defense and civilian nuclear research, to support activities which will benefit the United States as a whole. As such, this paper discusses the organizational details and requirements of the Neutron Source Reclamation Program at Los Alamos. This program has as its mission the retrieval, interim storage, and chemical reprocessing of ^{238}PuBe, ^{239}PuBe and ^{241}AmBe neutron sources residing in the hands of private companies and industries, academic institutions, and various state and Federal government agencies.

INTRODUCTION AND BACKGROUND

"The Los Alamos National Laboratory is dedicated to supporting United States Department of Energy's (DOE's) goals through the application of world-class science and technology to the maintenance of the nation's security and well-being. Los Alamos will continue its special role in defense, particularly in nuclear weapons technology, and will increasingly use its multidisciplinary capabilities to solve important civilian problems."(1) This quote is the opening paragraph for the Los Alamos National Laboratory's mission statement. One of the important overriding themes of the Los Alamos National Laboratory as a world-class institution is to utilize its expertise in enhancing the long-term welfare of society by minimizing negative side effects of nuclear technology over the past decades. The Los Alamos National Laboratory is therefore committed to the use of its technical competencies and nuclear facilities, developed through programs in the areas of defense and civilian research, to support activities which will benefit the nation as a whole.

Sealed sources are the most widely available type of nuclear material to the public in the United States (The United States Nuclear Regulatory Commission (NRC) estimates that there are nearly 27,000 sealed sources of all types that could eventually become Greater Than Class C Low Level Waste (GTCC LLW). About 40% of all sealed sources available for GTCC LLW status involve the radio nuclides plutonium and americium.). These sealed sources are small, encapsulated, relatively high radioactive materials with activities ranging between 1mCi to greater than 1,000 Ci. These materials have widespread applications in the areas of medicine, construction, manufacturing, and research. Examples of sealed source containing devices include calibration sources used for the calibration of radiation measuring and monitoring instruments; medical sources for the diagnosis and treatment of diseases using x-ray imaging, brachytherapy, and teletherapy; well logging devices for use in geophysical surveys; portable moisture and density gauges used at construction sites and on farms; and irradiation devices used in research, food processing, medical applications for irradiating blood, biological samples, etc.; fixed gauges used for monitoring of manufacturing production processes; and general neutron application devices used in a variety of industrial applications such as explosives detection and neutron activation analysis.(2)

CURRENT OPTIONS FOR DEALING WITH GTCC LLW SEALED SOURCES

Within the United States, the Low-Level Radioactive Waste Policy Amendments Act of 1985 gives responsibility for disposing of GTCC LLW to the United States Department of Energy. At present, the following options are available for ultimate disposition of plutonium and americium neutron sources:

Disposition Option 1

Do nothing; leave sealed sources in the hands of the public until an interim storage facility and GTCC LLW disposal capability are in place.

The consequences of this option include: 1) increased frequency of abandoned sources, 2) risk of contaminating the public due to a defective or breached source, 3) potential radiological health threat to the public, 4) theft or diversion for illicit purposed, radiological sabotage.

Disposition Option 2

Interim and long time storage followed by near-surface, intermediate depth, or deep geologic disposal.

The consequences of this option revert to those of Option 1 above for the immediate future (15-20 years). For long term storage, there are extensive costs and increased risks associated with extended storage, packaging for storage and repackaging for disposal. Disposal of plutonium would probably require a Category I Special Nuclear Material (SNM) storage facility and disposal site.

Total current dollar costs for development and operation associated with combined disposal of all GTCC LLW have recently been estimated to be $290-$630 M.(3) Total current dollar costs for closure and post-closure of a disposal site have recently been estimated to be $1.4-$2.0 B.(3) These costs will have to be incurred even if all Pu and Am sources are eliminated from the GTCC LLW category, unless other cost effective options are available to deal with other GTCC LLW forms.

Disposition Option 3

Eliminate Pu and Am sources from the potential GTCC LLW category by dismantling and separating the Pu and Am from the Be. Stabilize, consolidate and store the separated actinide material in an existing Category I SNM vault at Los Alamos.

The consequences of this disposition option are that "abandoned" sources can be dealt with in a timely manner. Health and safety risks and threats are also decreased by immediate removal of unwanted sources from the public domain. Appropriate regulatory permitting is required, glove box and existing hot cell modifications are required, and there is no impact on existing Los Alamos SNM storage vault.

LOS ALAMOS NATIONAL LABORATORY NUCLEAR MATERIAL PROGRAM

Over the past fifty years, the Los Alamos National Laboratory has developed an extensive capability to handle significant quantities of nuclear materials as part of its role in support of DOE Defense Program activities. The goals and objectives of these activities are met through a base program in nuclear materials technology research, development, and demonstration as well as through additional programs (such as the Neutron Source Reclamation Program) aimed at

specific, often near-term goals. This base program encompasses (1) plutonium process technology research, development and demonstration activities, (2) technology transfer and off-site technical support, (3) nuclear materials recycle and recovery, and (4) nuclear facility operations and maintenance.

Los Alamos National Laboratory Nuclear Facilities

Two major Los Alamos National Laboratory nuclear facilities of particular importance to the Neutron Source Reclamation Program are the TA-55 Plutonium Facility and the Chemistry and Metallurgy Research Building (CMR Building).

The TA-55 Plutonium Facility

The TA-55 Plutonium Facility is used to perform basic research on nuclear materials; technology development, demonstration and exchange, and to provide support for the DOE's nuclear materials missions. Major activities occurring within this facility include (1) research and development to study the fundamental chemistry and metallurgy of nuclear materials including most of the major actinide isotopes (^{238}Pu, ^{239}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{237}Np); (2) Development and Demonstration of enhanced technologies for all aspects of nuclear materials processing for the DOE Complex; and (3) Technology transfer and processing support which in addition to ^{239}Pu related activities also includes activities associated with Radioisotope Generator development using ^{238}Pu, and development of nuclear reactor fuel using ^{235}U.

Recovery activities involving plutonium-239 and americium neutron sources center around the use of hydrochloric acid as the basis for the process chemistry. These aqueous chloride processing activities require corrosion-resistant glove boxes operations which are available in the TA-55 Plutonium Facility, and which provide the only chloride processing chemistry operations for plutonium within the United States.

The Chemistry and Metallurgy Research (CMR) Building

The CMR building was designed to house research and experimental facilities for analytical chemistry, plutonium metallurgy, uranium chemistry, engineering design, and other support functions. At present all of the analytical chemistry capability to support Los Alamos nuclear materials programs are located in the CMR Building.

Hot cell facilities are also located within the CMR Building. Sixteen hot cells made of 28-inch thick concrete/magnetite walls are equipped with Lucite shields and remote manipulators to allow for handling, storing, and processing of highly radioactive materials of all types and quantities, including $^{238}PuBe$ and $^{241}AmBe$ neutron sources.

PURPOSE OF THE NEUTRON SOURCE RECLAMATION PROGRAM

The major purpose of the Neutron Source Reclamation Program is to lessen the nuclear danger by retrieving unwanted sources from the general public and various Government agencies, and to chemically process the sources in order to reduce the radiological risks associated with neutron radiation from α-n reactions and from the gamma radiation associated with ^{241}Am. Other reasons for processing neutron sources rather than disposing of them by burial include the excessive costs for burial, along with the fact that burial is environmentally unacceptable. The nation should be working from a materials management philosophy that minimizes the quantity of actinides being placed in the environment. Furthermore, consolidation of sources in

intermediate or long term storage vaults is not readily feasible as there is currently no such vault available nor will there be one available in the indefinite future.

HISTORY OF NEUTRON SOURCE RECLAMATION AT LOS ALAMOS

Since the mid 1950s the plutonium-beryllium alloy $^{239}PuBe_{13}$ has been used as a neutron source in the nuclear industry and in experimental laboratories. Over the years there have been approximately 17 different commercial manufacturers of these sources with over 2,500 sources manufactured in the United States.

When a source is no longer needed, it must be decommissioned and the plutonium recovered. To accomplish this handling, storage, and processing of the plutonium-beryllium neutron sources require very specialized facilities.(3) The Los Alamos National Laboratory has processed over 700 $^{239}PuBe_{13}$ neutron sources between 1979 and 1990 at the TA-55 Plutonium Facility. In June of 1990 the neutron source recovery line was modified to perform process development activities, and sources have not been processed at Los Alamos since that time.

The most recent commitment by the DOE to assist the public with unwanted neutron sources came in April 1991 when the Assistant Secretary for Defense Programs initially approved a $^{239}PuBe$ neutron source recovery program at the Los Alamos National Laboratory. The purpose of this program was for Los Alamos to accept $^{239}PuBe$ neutron sources from educational and other Government institutions over a three to four year period in order to process the sources and reduce the radiation exposure problems associated with these materials. Just recently Los Alamos has started working with DOE's Office of the Deputy Assistant Secretary-Waste Management to establish a program to retrieve and recover $^{238}PuBe$ and $^{241}AmBe$ sealed neutron sources from civilian and other Government organizations.

MAJOR PROGRAM ELEMENTS

Neutron Source Recovery Program Databases

Los Alamos is compiling information pertaining to $^{239}PuBe$ $^{238}PuBe$, and $^{241}AmBe$ sources. Using information obtained from DOE/Idaho, EG&G /Idaho, and other sources, three data bases have been constructed. The first database contains sources located off-site, that will eventually be shipped to Los Alamos for processing. The second contains a listing of sources presently residing at Los Alamos awaiting processing. The third database is an up-to-date mailing/contact list of institution which maintain control of $^{239}PuBe$, $^{238}PuBe$, and $^{241}AmBe$ sources.

Shipping Protocol and Coordination

As a courtesy to the shipping organizations, Los Alamos National Laboratory supplies the shipper with a shipping packet which contains detailed packaging procedures, monitoring procedures, shipping procedures, placards/labels, and all required shipping papers and forms. Because of the stringent requirements for shipping nuclear material, Los Alamos works closely with facilities which will be shipping sources to assure that the facility is in compliance with all regulations governing nuclear material shipments.

Interim Storage

The sources that are shipped to Los Alamos are placed in interim storage at the Plutonium Facility to create a feed queue for reprocessing operations. The Los Alamos Plutonium Facility will not store more than 200 sources at any one time. Additional interim storage capacity of up to approximately 1,000 sealed sources may be made available in the CMR Building hot cell area sometime during early 1996.

Chemical Processing

Upon verification of serial number, a neutron source is dismantled by first breaching the outer cladding. The actinide material (an oxide or beryllium alloy, depending on the source type) is then dissolved in hydrochloric acid, and subsequently separated from the beryllium using ion exchange techniques. The plutonium or americium is then eluted from the ion exchange column and precipitated from solution. Finally, the actinide precipitate is converted to plutonium oxide which is stored for possible future use. The beryllium is appropriately discarded as waste. Details of the ^{239}PuBe neutron source processing chemistry are described in Reference 4.

THE FUTURE FOR SEALED SOURCE RECLAMATION AT LOS ALAMOS

Within the United States there is currently no avenue for the removal and destruction of neutron sources other than those containing ^{239}Pu as discussed above. However, there have been demands from agencies both inside and outside the Federal Government as well as from the public to dispose of unwanted neutron sources containing ^{238}Pu and ^{241}Am. The Conference of Radiation Control Program Directors, Inc. (CRCPD) has specifically identified more than 1000 curies each of unwanted ^{239}PuBe and ^{241}AmBe sources in storage throughout the United States. In addition, the CRCPD has been advised by brokers that there are kilocuries of these unwanted materials in the hands of additional owners. Because of the significant interest in retrieving these neutron sources from the public domain, Los Alamos is working in conjunction with the DOE to implement a recovery initiative for the ^{238}PuBe and ^{241}AmBe sources . While the start of this new initiative is uncertain at the present time, we are attempting to obtain resources to start initial phases of a recovery program in 1995, and to begin accepting shipments sometime in 1996.

SUMMARY AND CONCLUSIONS

Sealed neutron sources containing ^{239}Pu are currently being dispositioned at Los Alamos as a service to the nation.. Experts in the industry indicate that managing unwanted ^{238}PuBe and ^{241}AmBe neutron sources residing in the public domain is going to be major problem unless a disposition option is available soon. Los Alamos has an existing ^{239}PuBe reclamation program, facilities, interim storage capacity, and technical staff with the relevant experience and knowledge in managing and processing neutron sources. This enables rapid extension of the current Los Alamos neutron source reclamation program to receive and reprocess ^{238}PuBe and ^{241}AmBe sealed sources in a timely fashion. No other DOE facility has the capability to deal with these sources in the indefinite future.

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