MANAGEMENT OF HEAVY ISOTOPES IN THE DOE COMPLEX

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ABSTRACT

Currently each Department Of Energy (DOE) Program office manages its own nuclear materials through activities such as production, processing, storage, transportation, and disposition. However, recognizing the need to strengthen its strategic approach to the integrated life-cycle management of nuclear materials, DOE established the Nuclear Materials Management Stewardship Initiative (NMMSI) in January 2000. The NMMSI's first visible product was the Integrated Nuclear Material Management Plan in which it was generally recommended that DOE take a cross-cutting look at managing its nuclear materials, and specifically recommended that four Nuclear Material Management Groups (NMMGs) be formed. These groups were established to facilitate management of nuclear materials for which DOE has or may have responsibility, including many presently not in DOE's direct control.

One of these NMMGs, the Heavy Isotope Management Group (HIMG) was established at Oak Ridge National Laboratory in December 2000, to facilitate management of (a) actinide and their decay products (except sealed sources) and (b) isotopically enriched stable and radioactive isotopes except uranium and lithium, but excluding thorium, uranium, spent fuel, and weapons or reactor-grade plutonium which are addressed by other NMMGs. Despite its short duration and relatively limited funding, the HIMG has facilitated the disposition of heavy isotopes from Lawrence Berkely National Laboratory (LBNL), Rocky Flats Environmental Technology Site (RFETS), Pacific Northwest National Laboratory (PNNL), Oak Ridge National Laboratory (ORNL), Lawrence Livermore National Laboratory (LLNL), and Idaho National Engineering and Environmental Laboratory (INEEL). The primary disposition options have been to facilitate reuse of valuable heavy isotopes by matching custodians of unwanted materials with other users that seek such materials for new applications. This approach has the dual advantages of avoiding custodian disposal costs plus cost to the user of obtaining newly produced material. The HIMG has also prepared issue papers on neptunium and americium/curium that identify the resources, potential uses, and disposal pathways for the materials across the DOE Complex. In the future the HIMG expects to comprehensively identify the status of the U.S. heavy isotope inventory, prepare additional issue papers and plans charting the future of this inventory, and to facilitate execution of the plan.

INTRODUCTION

Within the DOE the various Program Offices have specific responsibilities for certain nuclear materials and their uses. Since each Program has different needs and objectives, the evolution has naturally been for the Programs to take responsibility only for those under its direct control: to ensure needed supplies, storage, security, etc., to meet the projected programmatic needs. The Department recognized that the changing times and missions, influenced in no small measure by the downsizing of the Complex and the increased incentive to dispose of excess nuclear materials, now demands a more integrated approach to the management of nuclear materials.

Beginning in November 1999 the Task Force held workshops involving a few dozen nuclear materials and materials management experts from across the country to develop the framework for the integrated management of all the DOE nuclear materials. One that looks more at the management and overall uses of each material - no matter its use or "owner" - to help determine what is a valuable resource to retain and what should be disposed. The result of this effort was the completion of an Integrated Nuclear Materials Management Plan reported to Congress in June 2000. This plan committed the DOE to evaluate the option of establishing several nuclear material management groups to help maintain a core level of technical expertise and facility processing capability and to facilitate integration.

BACKGROUND

For more than 30 years the U. S. Government has utilized Americium and Curium as feed materials for the production of radioisotopes for research, medical, and industrial applications. ORNL has been a key facility in the production and distribution of these heavy elements during this time through the DOE Heavy Element program. Historically, the feed materials were supplied by the Savannah River Site and transported to ORNL. ORNL purifies the materials, prepares targets for irradiation, and processes the irradiated targets to produce a variety of materials.

The Radiochemical Engineering Development Center at ORNL processes Americium and Curium targets and typically has produced isotopes of Berkelium, Einsteinium, and Fermium for shipment to other laboratories to be used for research activities. Some of these materials are then used to produce the heaviest elements in the periodic table. Californium is also produced, separated, and manufactured into tailored sources for a wide range of industrial and government applications. There are now more than 100 formal loan agreements in force with over 60 institutions for hundreds of neutron sources containing ^{252}Cf.

The mainline products ultimately derived from the ^{243}Am and ^{244}Cm resources for loan and sale are ^{243}Am, ^{244}Cm, ^{249}Bk, ^{252}Cf, ^{253}Es, and ^{257}Fm. These are routinely provided to Argonne National Laboratory, Los Alamos National Laboratory (LANL), LBNL, LLNL, the Oak Ridge Associated

Universities, and occasionally to other laboratories in the US and overseas. The same is true for the derived products ^{248}Cm, ^{249}Cf, ^{254}Es, and ^{255}Fm. More specialized products, such as ^{240}Pu, ^{242}Cm, and other isotopes of californium are also routinely produced. From the "raw" materials, Americium-243 and Curium-244, more than 1800 shipments of trans-plutonium isotopes have been made.

The Heavy Isotope Management Group consists of a small core group of nuclear materials experts with special expertise and interest in the heavy elements. It was established as part of the Nuclear Material Strategic Initiative to provide leadership for an integrated approach that spans the complex for the management of these resources. The existing missions and the expertise available make Oak Ridge the natural location for the nexus of the HIMG. Also as part of the group are other experts and specialists located at other sites and laboratories across the country; resources available to support the Department's heavy isotope missions. Sites actively participating with expertise include Savannah River, Idaho, Richland, LLNL, LBNL, and LANL.

HEAVY ISOTOPES

The Heavy Elements are generally considered to be those with atomic number greater than 89 (Actinium). However, for the scope of activities of the HIMG there are some exceptions and caveats. Excluded are ^{230}Th and ^{232}Th (which are handled by the UMG), but not the other isotopes of thorium. Uranium is the purview of the UMG, except generally for the ultra high purity ^{235}U and some shared interests on ^{233}U. Likewise plutonium comes under the PMG, but because of special uses the isotopes ^{238}Pu, ^{240}Pu, ^{242}Pu, and ^{244}Pu are usually the concern of the HIMG.

The materials encompassed by the HIMG include Np, Cm, Am, ^{233}U, and others that could not be produced in quantity with any currently existing facilities. They are in many ways a national resource. Although the known uses for most are not large, there is always the potential for a new use; which cannot easily be anticipated and therefore used as justification for retention of the materials. The existence of most is a result of long irradiation of target materials in special reactors; reactors that are now shut down and will not be re-started. Nor is it anticipated that reactors of similar capability will be built in the foreseeable future.

Uranium

A case in point for unanticipated value is the most significant of the special uranium isotopes, ^{233}U. This isotope was produced during the 1960s and 1970s as part of the development of the thorium fuel cycle. Significant inventories of separated ^{233}U, mostly in the form of oxides, are stored at ORNL. A second major inventory, mostly in the form of fabricated fuel elements, is stored at INEEL. Small holdings of ^{233}U exist at LANL, LLNL, and a number of other sites. The primary decay product of ^{233}U is ^{229}Th. Through a series of decays the thorium-229 eventually becomes actinium-225, which is showing great promise in clinical trials for the treatment of cancer. When tagged onto a monoclonal antibody the actinium-225 decays to deliver very high doses of alpha radiation directly to the cancer with very little impact on healthy cells. Such a use could not have been anticipated even a few years ago and, had the "useless" uranium-233 been permanently disposed, there would now be no source of the material.

Another special uranium isotope tracked by the HIMG is ^{235}U isotopically-enriched to greater than 99.6%. This material has been produced in the calutrons operated by the isotopes program in Oak Ridge. Small quantities of calutron-enriched ^{235}U have been distributed throughout the nation, and the material continues to have applications in research and dosimetry. The third special uranium isotope is high-purity ^{234}U, separated as the daughter product of ^{238}Pu. The parent isotope is tracked by the HIMG as a special plutonium isotope.

Neptunium

Neptunium is recovered from irradiated fuels or targets and is essentially mono-isotopic, neptunium-237. While relatively small amounts have been used in national security applications, essentially all the recovered Np serves as a feedstock for the production of ^{238}Pu. The production of ^{238}Pu necessarily results in the gradual consumption of the Np feedstock, much as creating ^{239}Pu consumes ^{238}U.

Plutonium

The large quantities of weapons-grade and reactor-grade plutonium, consisting mostly of ^{239}Pu with varying quantities of ^{240}Pu and lesser quantities of ^{238}Pu and other isotopes, is tracked by the plutonium materials management group. As with thorium and uranium, several special plutonium isotopes of relatively high isotopic purity are tracked by the HIMG. In addition, bulk quantities of "heavy" plutonium (containing significant fractions of ^{242}Pu) that might serve as feed material for the heavy isotopes production program may be tracked by the HIMG on an individual-case basis. Plutonium-238 is used as a heat source for spacecraft power and heating systems and several other applications. High-grade ^{238}Pu is produced by the irradiation of ^{237}Np in a high-flux reactor such as the High Flux Isotope Reactor in Oak Ridge or the Advanced Test Reactor in Idaho. Separated ^{238}Pu materials not already part of the power program are tracked by the HIMG. Plutonium-242 has been used as a feed for production of heavy isotopes and for a variety of other applications. As noted above, bulk quantities of mixed plutonium may also be tracked by the HIMG as potential feedstock for heavy isotope production programs. Plutonium-244 is used as a standard for isotopic dilution mass spectrometry, and has other applications in the nuclear safeguards arena.

Americium

The principle and most visible application of ^{241}Am is its use as an ionization source in smoke detectors used in homes and other buildings, where one gram of ^{241}Am can make about 5000 smoke detectors. It has been used industrially in conjunction with a light element such as beryllium to make (α,n) neutron sources that are used in testing machinery and equipment, and for oil well logging. It has also been used as a gamma source to measure the thickness of glass during manufacturing.

Californium

Most isotopes heavier than curium-244 have such short half lives as to be useful only for research. An exception is californium-252 which has an extraordinary number of uses of high importance. The existence of such applications is a result of ^{252}Cf having (a) a half life of 2.64y, which is long enough to allow it to be produced and distributed but short enough for it to represent an intense radiation source, and (b) a decay mode that produces almost 4 neutrons per fission event. This extraordinarily high spontaneous fission rate results in ^{252}Cf continuously producing 2.3 million neutrons per second per microgram of material. The intensity of the neutron source from such a small amount of material has resulted in ^{252}Cf being used in dozens of applications, where its use saves lives, time, and money. The most important applications of ^{252}Cf are as reactor startup sources for almost all power reactors, for neutron activation analysis which can detect trace quantities of many elements, detection of non-metallic land mines, detection of explosives, neutron radiography (to detect flaws in parts, or even in entire aircraft) on-line analysis of bulk materials, and in medical application for treatment of cancers.

Curium

Curium has some application in research and as heat sources but its single most important use is as feed material for targets to produce californium and heavier elements.

MISSION AND GOALS

The HIMG provides an integrated planning resource that is particularly valuable to small sites and sites being closed. The closure sites continue to identify old sources and other materials that would be disposed unless re-used. Having the HIMG provides a single source for information, packaging assistance, and identifying re-use potential. With the decline of the nuclear mission it has become more difficult to locate special knowledge and assistance. Formation of this group has resulted in an association of experts who can be essential to reducing wastes by matching those with Heavy Isotope (HI) problems to those with needs. The HIMG provides that "who to call" first point of contact for help. Equally important is the enhanced opportunity for re-use. The HIMG is in position to match up surplus materials with needs. This function will frequently reduce the need for disposal and can show significant cost avoidance, especially to the Environmental Management (EM) and the Nuclear Energy (NE) programs.

The group has participated in visits to various DOE sites and has been able to offer pathways for disposition of special heavy isotopes in almost all cases. These actions alone have the potential to save millions of dollars. But the mission of the HIMG is broader than direct assistance to closure sites. Some local governments are very anxious that the DOE reduce its inventory of radioactive materials at facilities in their area. In fact, the Department has in some cases agreed to limits on those quantities. The HIMG has been the key to finding a safe "home" for some of the HI's at reasonable cost; sometimes providing "headroom" necessary to allow other materials to be brought in for important new programs.

There is currently no data base or set of data bases that effectively identify heavy isotope "inventories." As knowledge of holdings improves, having the HIMG offers the opportunity to create that inventory; thus creating and maintaining a knowledge of forms, quantities, and locations of nuclear materials. As can be seen from the number of uses already known for these materials, they are already a national resource in very short supply. Perhaps more importantly, they are virtually irreplaceable.

The HIMG has as one key role to participate with the other Groups to assist in carrying out the NMMSI, an initiative we believe to be critical to the effective long term management of nuclear materials in this country, if not the world. This includes active involvement in the enabling documents, and technical contributions to long range planning and technology development. The HIMG acts as the coordinator of expertise for heavy isotope use and processing, as well as being a research and development center.

One of the more important functions of the Group is to provide expert knowledge and advice on heavy isotopes as national resource materials. As previously stated, the heavy isotopes are a precious commodity in limited supply. They were produced in very large, high flux reactors after very long irradiation times. To build new reactors and facilities to produce more of the heavy isotopes would cost at least tens of billions of dollars. There is a national program to develop Accelerator Transmutation of Waste to remove much of the nuclear hazard from spent nuclear fuel. This program requires hundreds of grams of Am, Cm, and Np for research and development efforts before it can be brought on line. The program is part of NE but most of the holdings are possessed by EM. The HIMG has been in the lead to highlight this need and to champion the retention of material streams that contain the needed isotopes.

Plutonium-244 is of critical importance in nuclear non-proliferation efforts and arms control. The HIMG is part of a national team to find a path for recovering and purifying this material from probably the only remaining source in the United States. HI expertise is critical to this kind of analysis and to developing the necessary systems to refine the material.

Some of the long term goals of the HIMG are:

- Review and revise the national inventory information on isotopes, characterization information, packaging, and storage locations and conditions.
- Act as a clearinghouse for identification of (1) materials to be stored, (2) transportation requirements, (3) stable forms for storage, and (4) potential storage locations.
- Be the coordinator of expertise for processing knowledge and the responsible group to provide assistance to sites where unique, or isolated, amounts of heavy elements are located.
- Support the beneficial use and commercial production of heavy isotopes in developing markets through technical expertise and assistance to others.
- Perform assistance for day-to-day planning, control, and execution of heavy isotope activities.
- Establish a management system for defining and projecting infrastructure needs:
    - Identify or develop adequate storage facilities;

  - o Maintain or provide adequate equipment necessary to process handle, or store heavy isotope materials;
  - o Develop disposition plans for materials not meeting current waste acceptance criteria and;
  - o Identify or acquire adequate certified shipping containers for assigned materials.
- Maintain advanced technical knowledge and capability for heavy isotopes:
  - o Develop and maintain necessary technical standards in such areas as handling, shipping, and storage;
  - o Establish and maintain partnerships with national laboratories, universities, and industry to improve the sciences for heavy isotope materials and;
  - o Facilitate the de-inventory of heavy isotope materials from closure sites to support mortgage reduction.

CONCLUSION

The HIMG has grown out of the DOE Heavy Element program and the DOE NMMSI. This program, with ORNL as the producer of heavy isotopes from available feedstocks, has supplied those materials to researchers and industry for over 30 years. During that time a nationwide network of experts with special knowledge of these nuclear materials evolved. However, with the steady decline of nuclear programs in the United States over the last 20 years, there has been a gradual loss of talent and history to retirements and career changes.

Today this deep expertise is thinned and scattered across the complex and is slipping away at an alarming rate. The HIMG is a "virtual group" of these experts with the knowledge to assist the DOE in its effective management of the heavy isotopes. It is the "who to call" for sites with these materials as problems for which they do not have an answer. There are some of these isotopes that should be viewed as national resources, and preserved for future use. The HIMG contributions to date related to uranium-233, plutonium-244, the ^{238}Pu production program, the ATW program, and site assistance at Berkely, Richland, Rocky Flats, and others have returned huge benefits for a very small investment. As sites are cleaned up and closed it is imperative that low cost and effective means of disposal are utilized, but equally important that the unique nature of them is recognized and managed for the future. Working with the other Material Management Groups, the DOE Program Offices, and companies across the nation, this team can help to accomplish the goals of the Strategic Initiative.