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WASTE MINIMIZATION IN THE LOS ALAMOS MEDICAL RADIOISOTOPE
PROGRAM

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ABSTRACT

Since the mid-1970s the Los Alamos Medical Radioisotope Program has been irradiating target materials to produce and recover radioisotopes for applications in medicine, environmental science, biology, physics, materials research, and other disciplines where radiotracers find utility. By necessity, the chemical processing of targets and the isolation of radioisotopes generates radioactive waste materials. Recent years have brought pressure to discontinue the use of hazardous materials and to minimize radioactive waste volumes. Substantial waste reduction measures have been introduced at the irradiation facility, in processing approaches, and even in the ways the product isotopes are supplied to users.

INTRODUCTION

For two decades the Medical Radioisotope Program at Los Alamos National Laboratory has been producing and recovering radioisotopes from targets irradiated at the Los Alamos Meson Physics Facility (LAMPF).¹ The 800 MeV proton beam with currents up to 1 milliAmpere provides unique opportunities in the field of radioisotope production. With these high currents and energies come many waste handling challenges. Equipment at the LAMPF irradiation facility is subjected to

extreme conditions resulting in the need to periodically replace and dispose of highly radioactive components. Spallation reactions result in a large number of isotopes associated with a diverse group of elements produced in each target.² The chemical recovery of individual radioisotopes requires specific and sometimes elaborate procedures resulting in the production of radioactive waste materials in the form of used reagents and labware. The shipment and subsequent return of materials used to supply purified radioisotopes to users result in contaminated lead shields and packaging material that must be cleaned or discarded. The laboratory has a radioactive waste disposal site which makes it possible to dispose of radioactive waste provided it is not contaminated with hazardous materials. Hazardous materials include carcinogens, toxic metals, flammable solvents, etc. The laboratory does not have a facility to dispose of hazardous materials but relies on an outside contractor for this service. Mixed waste is a combination of radioactive and hazardous materials. At this time, neither the laboratory nor outside contractors have a mechanism to dispose of this type of material. In recent years we have been subject to more restrictive regulatory, financial, and moral directives to eliminate mixed waste and minimize hazardous waste materials. Consequently, as we have developed approaches and techniques to perform irradiations, process targets, and provide radioisotopes to users, we have also pursued ways to deal with the unique waste generated by this work.

METHODS

LAMPF Irradiation Facility

The LAMPF irradiation facility employs 9, twenty-six foot long target stringers.³ Each stringer supports a target in the proton beam of the accelerator and provides cooling

water to the target. Originally the core of each stringer was filled with concrete containing lead shot for radiation shielding. Over time, the program has had to replace some of these stringers due to mechanical failures caused by the extreme operating environment. Because lead is a hazardous material these stringers were considered a form of mixed waste. The lead shot in the core has been replaced with magnetite, making them no longer a mixed waste when replacement is required. Engineering changes including bearings, welded water line fittings, and alignment fixtures have extended the life of new stringers thus reducing the contribution to the total volume of radioactive waste. In addition, the program now removes and reuses the portion of the stringer that is not highly activated thus reducing the associated waste even more.

Video cameras are used to monitor remote loading and removal of targets at the irradiation facility. In the past, these cameras have required replacement at frequent intervals due to failure caused by the high radiation environment. Shielded enclosures with leaded glass windows have greatly extended the life of these cameras and protected them from contamination and activation reducing this contribution to the radioactive waste stream.

Modifications to the remote handling tools used for the loading and removal of targets have made it possible to replace damaged portions of the tooling without replacing the entire tool. Weaker shear pins are used to attach the portion used by the operator then the shear pins that attach fixtures that support targets and tighten bolts. If a tool is broken, the failure is not in the remote environment and the damaged portion is easy to replace.

The external target encapsulation, called a carrier, is made of aluminum and was initially bolted closed.⁴ The seals for this closure could not tolerate long irradiations, and thus carriers for such irradiations required frequent replacement. As a result of extensive testing of various

aluminum alloys and closure mechanisms, the program is now using forged 2219 aluminum alloy target encapsulation that is welded closed. This arrangement has made it unnecessary to replace encapsulation to perform extended target irradiations. Minimizing the size of the encapsulation has also reduced both the bulk and the amount of radioactivity associated with the carrier when it is discarded. The approach of being able to replace only a portion of a damaged piece of equipment, and engineering and materials improvements to extend the life of mechanisms has made a substantial reduction in the waste associated with the irradiation of target materials.

Discontinuation of Mixed Waste from Target Processing

A major objective has been the elimination of the production of mixed waste and the associated use of hazardous materials in target processing. Because of the wide variety of isotopes associated with the many elements produced by spallation in targets, recovery procedures present significant challenges requiring a host of chemical separation techniques. Unfortunately many traditional and some new and very powerful approaches employ hazardous materials. For example, solvent extraction procedures have been a common mechanism for performing radiochemical separations. Unfortunately many extractants, or the solvents used with them, have been found to be hazardous materials. Ion exchange procedures have often been enhanced by the incorporation of water miscible organic compounds. Many of these compounds are now considered hazardous. Gaseous isotope recovery procedures often have involved hazardous materials to make cold temperature baths to condense out components. Many reagent gases used in chemical procedures are now considered hazardous. Partially used cylinders of hazardous gases associated with radioactive contamination

have become extremely difficult mixed waste disposal problems.

The elimination of hazardous materials associated with the recovery of radioisotopes has posed significant challenges but not without some unexpected benefits. For example, early procedures for recovering aluminum-26 from potassium chloride targets employed an extraction step using acetylacetone.⁵ This hazardous compound is no longer used; instead a precipitation step is employed to separate the aluminum isotope from the target solution, followed by dissolution and classical ion exchange methods. This new procedure provides an extremely valuable radiotracer with no mixed waste generation and reduced waste volume as compare to our previous aluminum-26 production method.⁶ Acetylacetone was also used in an extraction step to isolate vanadium-48,49 and beryllium-7 from irradiated zinc oxide targets.⁷ An alternative procedure has been developed that employs a precipitation step followed by conventional ion exchange techniques to isolate these isotopes.

A particularly good example of our efforts to eliminate mixed waste is the processing of molybdenum targets. Molybdenum targets are processed principally for the recovery of Sr-82. Five or six of these targets are processed each year. Each target produces two to twelve curies of Sr-82 depending on the length of irradiation. For many years we used an ion exchange procedure that employed as much as 4.5 liters of dioxane to process each target.⁸ Elimination of the dioxane from the chemistry was accomplished by dissolving the molybdenum metal in hydrogen peroxide instead of a mixture of nitric and phosphoric acid.⁹ This change eliminated the mixed waste problem but the volume of waste increased from an average of 11.5 liters to 17.2 liters. This change also resulted in a more difficult target dissolution and precipitation problems associated with the dissolved molybdenum solution. Additional work resolved the

precipitation problems making it possible to decrease molybdenum solution volumes. Eliminating the phosphoric acid from procedure also made it possible to recover zirconium-88, a feature not possible using the original procedure.¹⁰ In addition, a new procedure was developed that utilized the improvements in the hydrogen peroxide dissolution, and further reduced waste volumes.¹¹ The new procedure also increased the Sr-82 yield and had the added benefit of making it possible to recover technetium, rubidium, selenium, arsenic, and germanium isotopes.^{12,13} The flow charts demonstrate the evolution of our Sr-82 recovery procedures. (Figures 1,2,3)

Minimization of Target Processing Waste Volume

Target processing waste is primarily composed of hot cell fixtures, plasticware, glassware, and reagents. Corrosion is the biggest contributor to the failure and subsequent disposal and replacement of hot cell hardware. Plasticware and glassware are used to introduce reagents to the hot cells and to perform chemical separations. The chemical processing of targets results in the distribution of different radionuclides in the used reagents. These reagents must be neutralized and converted to a solid form before disposal. The laboratory does not have a system for treating a liquid waste stream containing high levels of the isotopes produced in our targets. Therefore it is important to minimize liquid waste volumes.

The program's efforts to minimize the replacement of hot cell fixtures has focused on reducing chemical corrosion in the cells and a modular approach to equipment used. The use of spill trays, good housekeeping, and condensers during evaporation procedures are the most effective approaches in controlling chemical corrosion. These steps have greatly extended the life of hot plates, pH meters, radiation

detector probes, and other electrical appliances. In addition, it has reduced the frequency at which hot cells require refurbishment. This represents a substantial reduction in radioactive waste, since refurbishment requires disposal of the entire contents of the cell, decontamination, and replacement of all appliances and hardware. Using equipment with replaceable, modular parts in a remote environment minimizes down time and further reduces the volume of the waste stream.

Plasticware and glassware is saved and reused for the processing of additional targets and often for the subsequent recovery procedures of other product isotopes from the same targets. In doing this, some caution must be taken. Glass and plastic are damaged by high radiation fields. Losing a valuable target because of a questionable piece of glassware is no savings. Bulk associated with plasticware is primarily in the form of plastic bottles; many of these are reused as primary containers for neutralized and sorbed reagent waste. Used glassware is crushed to minimize its contribution to the waste bulk.

Neutralized and immobilized reagents represent the biggest contribution to the volume of radioactive waste generated by processing irradiated targets. Consequently the minimization of reagent volume has become an important concern in designing chemistry procedures. A significant reduction in reagent waste volumes from ion exchange procedures has come from using one ion exchange column to separate the bulk of target material from radionuclides of interest, then employing a second smaller column or series of smaller columns for the final isolation of the nuclides of interest. Past ion exchange procedures concentrated on using one large column to perform the complete separation. This approach resulted in substantially larger reagent volumes employed in the separation and recovery process. This

improvement has been especially useful in reducing waste volumes in procedures to recover cadmium-109 from indium targets and strontium-82 from molybdenum targets. An added benefit has been improved purity of these radioisotope products.

Other Major Sources of Waste

Materials brought into the wing of the facility where the hot cells are located are treated as if potentially contaminated due to the difficulty of proving otherwise. This means that all articles that have entered the area must be disposed of as some form of radioactive waste, whether they have truly been contaminated or not. In the past this represented a substantial volume of low level radioactive waste. All discarded packing materials, laboratory equipment, reagents, instruments, tools, and metal stock became part of the radioactive waste stream. Unpacking materials outside of the radiologically controlled area coupled with the policy of ordering quantities of materials in a "buy only what you need" manner has significantly reduced the volume of such waste.

Some material must be unpacked in a radiologically controlled area due to potential contamination of the articles. The greatest contributor of this part of the waste stream had been the packing associated with returned shields from radioisotope product shipments. This source of waste has been virtually eliminated by developing a non-returnable shipping system. When radioisotope shipments require a large returnable lead shield, a reusable high density polyethylene foam box liner minimizes the waste. Formerly a disposable liner was used. Previous shipping systems resulted in more than a cubic foot of waste associated with each returned shield and frequently required decontamination of the lead shield, thus generating mixed waste.

Future Goals

Future steps toward the goal of waste minimization will undoubtedly require more effort to accomplish smaller gains. The program is currently investigating compaction mechanisms for hot cell waste. A concern is that too high a concentration of radioactive material in each waste container may result in increased personnel exposure hazards. Allowing waste to decay before disposal may help alleviate this problem but takes up space in the limited area of the hot cell facility. A more attractive solution to this problem is to sell the waste. This can be done by recovering radioactive species that have potential markets. Accordingly we have developed procedures for recovering sodium-22 from target encapsulation, and technetium-95m, arsenic-73, selenium-75, and germanium-68 from molybdenum target solutions.

CONCLUSIONS

Substitution of non-hazardous materials has made it possible to continue to produce many needed radioisotope products without the generation of mixed waste. The program has found that chemical separation approaches such as precipitations can work just as well as organic extractants and are easily adapted to hot cell use. Ion exchange and electrochemical procedures have a variety of parameters that can be modified without the use of hazardous organic compounds. Radiation shielding made of materials other than lead requires more bulk for the same shielding but often have greater strength and requires less fabrication effort. We are successfully using magnetite, brass, copper, tantalum, tungsten, and steel, in place of lead.

Many approaches have been employed to minimize the volume of waste generated by the program. The concept of modifying chemical separation procedures to minimize reagent

waste volumes has substantially reduced the quantity of waste leaving the hot cells. Minimizing the introduction of materials into a radiologically controlled area has made a substantial impact on the volume of the waste stream with a minimum of effort. The evolution of designs and materials choices has extended the life of irradiation hardware and minimized it's contribution to our waste. These improvements, coupled with the approach of buying only what is needed and disposing of only the damaged portion of a piece of equipment have not just reduced the volume of waste, but have also provided financial savings. A significant part of our program now involves waste-minimization research and development. Future in-roads into this problem will be realized as this work matures.

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REFERENCES

1. H. A. O'Brien, P. M. Grant, A. E. Ogard, Prog. Nuc. Med., 4 (1978) 93.
2. G. Friedlander, J. W. Kennedy, E. S. Macias, J. M. Miller, Nuclear and Radiochemistry, John Wiley and Sons, NewYork, 1981.
3. C. E. Cummings, A. E. Ogard, R. H. Shaw, Proceedings of 26th Conference on Remote Systems Technology, (1978) 201.
4. G. E. Bentley, J. W. Barns, T. P. DeBusk, M. A. Ott, Proceedings of 26th Conference on Remote Systems Technology, (1978) 378.

5. F. J. Steinkruger, D. R. Phillips, "Production of Aluminum-26", U.S. Patent #5,057,269, Oct. 15, 1991.
6. K. W. Thomas, *Radiochim. Acta*, 33 (1983) 213.
7. K. E. Thomas, *Radiochim. Acta*, 39 (1986) 69.
8. K. E. Thomas, J. W. Barnes, *Radionuclide Generators, New Systems for Nuclear Medicine Applications*, F. F. Knapp and T. A. Butler, eds., American Chemical Society, Washington, DC., 1984.
9. K. E. Thomas, *Appl. Radiat. Isot.*, 38 (1987) 175.
10. K. E. Thomas, *Radiochim. Acta*, 37 (1984) 137.
11. R. C. Heaton, D. J. Jamriska, W. A. Taylor, "Process for Strontium Separation", U.S. Patent #5,167,938, Dec. 1, 1992.
12. D. R. Phillips, D. J. Jamriska, V. T. Hamilton, "Recovery of Germanium Isotopes from Irradiated Targets", Patent #5,190,735.
13. R. C. Heaton, D. J. Jamriska, W. A. Taylor, "Process for ^{88}Zr , ^{88}Y , and ^{83}Rb Separation from Molybdenum" Serial Number 07981449.

Procedure StepsWaste Volume

Dissolve molybdenum
55% HNO₃
25% H₃PO₄
20% H₂O

4 Liters

Dilute with equal
volume of dioxane

4 Liters

Cation column followed
by 250 mL 50/50
dioxane/H₂O

0.25 Liters

8.25 Liters Mixed Waste

Elute column
500 mL H₂O
500 mL 0.5 M NH₄Cl
750 mL 0.5 M α-HIB
100 mL H₂O
750 mL 0.5 M HCl
250 mL 6 M HCl

3.1 Liters

Fume with HNO₃

Convert to 0.6 M HNO₃
Load activity on a 5 mL
cation exchange column
followed by a 2 M HNO₃
wash to elute Sr-82

0.1 Liters

Convert Sr-82 to
0.1 M HCl for
product

3.2 Liters Radioactive Waste

11.5 Liters Total Waste Volume

Fig. 1 Original Sr-82 Procedure (300g Target)

<u>Procedure Steps</u>	<u>Waste Volume</u>
Dissolve molybdenum With H ₂ O ₂	13.5 Liters
Cation column followed by 10% H ₂ O ₂	0.5 Liters
Elute column 500 mL H ₂ O 500 mL 0.5 M NH ₄ Cl 750 mL 0.5 M α-HIB 100 mL H ₂ O 750 mL 0.5 M HCl 250 mL HCl	2.85 Liters
Fume with HNO ₃	0.25 Liters
Convert to 0.6 M HNO ₃ Load activity on a 5 mL cation exchange column followed by a 2 M HNO ₃ wash to elute Sr-82	0.1 Liters
Convert Sr-82 to 0.1 M HCl for product	
17.2 liters Radioactive Waste	
0 Liters Mixed Waste	

Fig. 2 H₂O₂ Sr-82 Procedure (300g Target)

Dissolve molybdenum with H_2O_2 , load on a cation exchange column	7.5 Liters
Wash column with 10% H_2O_2 , then H_2O	1.5 Liters
Strip activity with 6 M HCl	1.0 Liter
Load activity on 10 mL cation column, wash with 0.5 M H_2SO_4 and H_2O	0.4 Liters
Elute Sr-82 with 1 M HCl	0.5 Liters
Convert Sr-82 to 0.1 M HCl for product	

10.9 Liters Radioactive Waste
0 Liters Mixed Waste

Fig. 3 Revised H_2O_2 Sr-82 Procedure (300g Target)

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