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ABSTRACT

The isotope separation program at Oak Ridge National Laboratory (ORNL) prepares and distributes electromagnetically enriched stable isotopes to the worldwide scientific community. Among the topics discussed in the present paper are the methods of enriching isotopes, the limitations that apply to the quantity and final assay of the separation products, and a generalized production flowsheet indicating the capability of the facility. A brief description of each of the production steps, from the selection and preparation of initial feedstock to the recovery and distribution of the isotopically enriched material, is presented. The future of the facility, the continued supply of enriched isotopes, and the response of the program to new and changing requirements are emphasized.

INTRODUCTION

The existence of the electromagnetic isotope separation facility at the Oak Ridge National Laboratory (ORNL) and the availability of enriched stable isotopes from the Isotope Sales Office are familiar to most research and commercial laboratories throughout the world. The purpose of this paper is to review the goals of the program, the technology employed to produce the separated isotopes, and the restrictions and limitations that apply to both the quantity and isotopic purity achievable. The discussion will be limited to the approximately 60 multi-isotopic elements made available by the operation of the electromagnetic facility. The isotopic enrichment of the light elements, notably hydrogen, lithium, and boron, as well as the production of the isotopes of the gaseous elements which are available from the Mound Laboratory, will not be addressed in this presentation.

The objective of the ORNL isotope enrichment program is to *isotopically enhance and distribute stable isotopes, selected radioactive isotopes, and heavy-element isotopes for use in research and development and in commercial activities.* To accomplish this objective ORNL, at the direction of the Department of Energy (DOE), operates the electromagnetic isotope enrichment facility.

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This facility consists of very high current mass separators, "the calutrons," which were used to produce enriched uranium during the 1940's. An important aspect of the overall efforts is the research and development activities which are directed toward increasing both the throughput and isotopic purity of the products. Further, the program continually evaluates alternative enrichment techniques which would either supplement the electromagnetic method or serve as a more cost-effective technology. The distribution of the enriched stable isotopes is accomplished through the Isotope Sales Office, which is also located at ORNL. The distribution falls into two distinct categories: (1) a DOE-sponsored Research Materials Collection (RMC) from which multigram quantities of enriched materials are made available to the DOE research community on a no-cost loan basis for nondestructive research, and (2) the direct sale of enriched isotopes to the worldwide research and commercial communities on a cost-recovery basis.

THE ELECTROMAGNETIC ENRICHMENT FACILITY

Description

The calutron facility is a unique national asset, since the USSR is the only other nation possessing a similar capability. Many other nations have isotopic enhancement capabilities at their research laboratories; however, they are usually of limited size and scope and, in many instances, are utilized for specific purposes and programs.

There are two "tracks" comprising the facility with one track reserved for stable isotope enrichment and the second for radioactive and actinide elements. The magnetic field is horizontal but, rather than the original design of a single magnetic field linking all 36 separators, the stable isotope track has been subdivided into four segments. This segmentation has been accomplished by inserting magnetic shunts from one side of the track to the other. This configuration results in three banks of eight separators and one bank of six separators. By exciting the magnetic field in each segment independently, one can theoretically enrich the isotopes of four elements simultaneously in the thirty available calutrons.

All of the separators have their source and collectors in the magnetic field. The advantage of this configuration is that it allows a high degree of beam-charge neutralization, and thus the separator can maintain high-current densities without degrading the focal qualities. The disadvantage is that servicing the source or collector requires that the entire tank be let down to atmospheric pressure, and thus a fraction of time is spent in reestablishing high vacuum. Two basic types of separators are available for the enrichment of the stable isotopes. The first of these is the conventional, 61-cm radius, 180° focusing

calutron unit. The second is a 255° sector focusing device with the magnetic field index equal to 0.5. Six of the separators are equipped with magnetic polefaces to give the double focusing properties. These latter separators have a dispersion approximately 50% greater than the standard unit, and thus the higher theoretical mass resolution results in a product with greater isotopic enhancement. The throughput associated with these separators, however, is correspondingly less than that achieved with the standard calutron units. In actual practice, the 255° units are used when isotopic assay is the prime consideration and the 180° units are operated when maximum yield is desired. The actual number of units operated at any one time is dependent on the available DOE funding.

Capability

A brief review of the basic steps in electromagnetic isotope enrichment will help in establishing the capabilities of the facility. The feed material, in either elemental or compound form, is introduced into the calutron source where it is either heated and vaporized directly or heated in a stream of carbon tetrahalide to form a volatile halide. This vapor is then introduced into an arc discharge where it is ionized in the high-current source. The ionized particle is extracted from the ion source and accelerated to approximately 40 keV, bent in the magnetic field with a mean radius of curvature of 61 cm, and the individual isotopic beams intercepted by isotope collectors. The collectors (one for each isotope), which are constructed of carbon, copper, or aluminum, are located behind a slotted face plate. Following a run, whose duration may be between 50 and several hundred hours depending on the element, the collectors are removed from the separator and the material extracted from each individual receiver, chemically purified, assayed, and made available for inventory. Obviously, the relative quantity of enriched product in each mass fraction is directly proportional to the isotopic assay of the feed.

As might be expected, each element or compound has unique operating characteristics. Thus, making generalizations with regard to the throughput capability of the facility is difficult. The ion sources produce a beam whose dimensions are approximately 0.4 x 13 cm. Typical beam currents between 10 and 100 mA are obtained, with the average in the 25-50 mA region. As a rule of thumb, one separator can provide approximately 0.1 mol of an element per 24-hr operational day. As noted above, this figure must be multiplied by the natural isotopic abundance to determine the yield for a particular isotope.

It is considerably more difficult to present a similar universal rule for the achievable isotopic purity. Isotopic purity is strongly dependent on the isotope desired and the abundance of its nearest neighbors. In addition, the

vaporization and ionization characteristics of the element, the probability of the isotope remaining in the collector, and the degree of beam focus which can be maintained are all intimately related to the final product assay. However, in the spirit of this paper and with the above considerations clearly in mind, one may approximate the assay by applying a decontamination factor. The ratio of the final assay divided by the tails contamination to the initial assay divided by the feed contamination is given approximately by 23,000 divided by the mass of the isotope. Applying this, for example, in the lead region yields a decontamination factor on the order of 110. This would predict that an isotope with 5% initial abundance could be enriched to approximately 85% in a single pass with a standard 180° calutron.

The products from one separation may be recycled to obtain a significantly higher isotopic assay in a two-pass campaign. This concept is expensive since the process efficiency is significantly less than unity. The process efficiency is defined as the ratio of the quantity of material removed from the collectors to the quantity of charge material vaporized in the ion source. As one might expect, the process efficiency is a function of source performance and is typically between 5 and 25%, with the average being approximately 10%. The unresolved material can be recovered from the separator following an enrichment campaign. This is normal procedure when the feedstock is either enriched or of sufficient value to justify the cost of recovery.

The remaining capability within the facility is the direct production of isotopically enriched surface-deposited targets or the production of small quantities of very highly enriched material. The device used for the preparation is a 180° sector separator with the source and collector external to the magnetic field. The device has a magnetic field configuration resulting in a dispersion roughly seven times that of the 180° production calutron unit. Therefore, the achievable isotopic enrichment is significantly greater. However, the ion throughput is correspondingly lower. Thus, although the machine is useful for making special targets, it is only marginally efficient for producing multigram quantities of highly enriched isotopes. In a typical target fabrication, the ions are slowed to approximately 200 eV and allowed to impinge on a suitable backing such as carbon, aluminum, or gold foil. The material is deposited as a line image 1-2 cm x 2 mm, with thicknesses up to $\sim$100 $\mu g/cm^2$. The target shape can be enlarged by either oscillating the target or by defocusing the beam after the mass defining slit.

Research and Development

Many of the activities traditionally associated with basic research of the underlying principles of electromagnetic isotope separation are currently

directed into those areas more properly called process improvement. The following discussion will indicate some of the achievements of this effort.

The ion source region offers the greatest potential for immediate improvement. It is, however, the most complex area of the process. It is within a small volume of space that the variables associated with the feed vapor production, ionization arc current, plasma density, electrostatic and magnetic fields, and system pressure interact. The problem is, therefore, to isolate and optimize one of the variables. The experience with the 255° separator ion source illustrates the utility of this approach. By carefully tailoring the dimensions and curvature of the ion exit slit, both the beam current and the isotopic enrichment were significantly improved. The total tin current (product) increased by almost a factor of two and the isotopic assay of ^{122}Sn improved from ~95% to >98%. In a similar way, systematic investigation of feed compounds have yielded improvements in the production rate of ^{203}Tl. The use of alternative configurations of extraction and accelerating electrodes have permitted a reduction in the background pressure at the ion source, and thus in beams with a higher degree of focus.

Emphasis is also placed on the design and materials used in the ion collectors. A new innovation has been employed in the enrichment of tellurium. A specially designed water-cooled aluminum receiver assembly was used which incorporated "skimmers" to limit the angular acceptance of the pocket. The resulting assay of ^{124}Te showed that unwanted isotopic contamination could be reduced by as much as a factor of two.

INVENTORY AND DISTRIBUTION

As is noted in the Introduction, the material produced in the facility is allocated to two independent inventories. The first is the DOE Research Materials Collection. The current DOE policy regarding circulation of the RMC is that this material is available on loan to U.S. scientists for use in nondestructive experiments. This material can also be used in experimental facilities outside the United States, but with restrictions. Among the stipulated conditions are that the experiment must be of relevance to the DOE mission and that the research must be a collaborative effort with a U.S. scientist who has assumed responsibility for the integrity of the sample. There are certain exceptions to this policy, for example, the use of RMC samples within the European community for the study of neutron cross sections. In this case, the samples are arranged for and placed in the custody of EURATOM. Such loans are usually made only at the strong recommendation of the European-American Nuclear Data Committee (EANDC).

The sales inventory, on the other hand, is available to the worldwide community. Distribution is made to laboratories engaged in nuclear medical, commercial, and basic research activities. The material is sold on a first-come, first-served basis. Thus, although one may obtain a quotation of price and availability of material, a sample is committed only upon receipt of a valid purchase order. This can cause confusion and, in some cases, result in dissatisfaction with the sales operation. The relative needs of the various market segments served by the sales inventory present an interesting picture. The research community utilizes relatively small amounts of almost all the enriched isotopes. The nuclear medical and industrial sectors require large quantities but of a very limited number of isotopes. Further, there are some isotopes of many elements which are not wanted by anyone. The total demand is always factored into the production schedule.

The current operation is such that the inventory is being depleted at a rate greater than it is being replenished. At the present time, approximately 65 isotopes are totally depleted from the sales inventory. With a few notable exceptions these isotopes are needed almost exclusively for research purposes. The isotopes which are being impacted most heavily at present are those that are in high demand (i.e., used by the industrial and nuclear medical segments). Our main concern is to maintain a viable stock of these isotopes. To accomplish this with a limited-scope operation will mean that other items in the overall inventory will be totally depleted for some period of time. Special separations of relatively small quantities of material can be accommodated within the facility; however, the cost of materials produced by this mechanism is greater than that for material obtained through the normal production channels. From our viewpoint, the inventory size and associated cost are prime concerns. One must balance an efficient mode of production with the anticipated demand for a particular isotope. The response time to provide material in a normal production mode is on the order of 6 to 8 months. This is due to the long lead time required for constructing equipment, preparing for the separation of a particular element, recovery of the material, and processing it to a chemially pure product.

SUMMARY

This paper has attempted to present a brief description of the production steps, from the selection and preparation of the initial feedstock to the recovery and distribution of the isotopically enriched materials. The facility suffers from the disadvantage of coping with utility and support systems that are rapidly becoming obsolescent and that the current operational level is insufficient to maintain sales inventory equilibrium. The electromagnetic isotope enrichment facility does, however, have the operational equipment and

capability to almost triple the current production level. Doubling the number of separators currently in operation, that is, utilizing the full complement of 30 calutrons in the stable isotope track, offers the potential of maintaining inventory equilibrium and permitting the gradual reinstatement of the depleted isotopes. This increased production can be achieved as rapidly as an expanded operational crew can be trained.