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**STATUS OF STABLE ISOTOPE ENRICHMENT, PRODUCTS, AND  
SERVICES AT THE OAK RIDGE NATIONAL LABORATORY**

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# STATUS OF STABLE ISOTOPE ENRICHMENT, PRODUCTS, AND SERVICES AT THE OAK RIDGE NATIONAL LABORATORY

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## Abstract

The Oak Ridge National Laboratory (ORNL) has been supplying enriched stable and radioactive isotopes to the research, medical, and industrial communities for over 50 years. Very significant changes have occurred in this effort over the past several years, and, while many of these changes have had a negative impact on the availability of enriched isotopes, more recent developments are actually improving the situation for both the users and the producers of enriched isotopes. ORNL is still a major producer and distributor of radioisotopes, but future isotope enrichment operations conducted at the Isotope Enrichment Facility (IEF) will be limited to stable isotopes. Among the positive changes in the enriched stable isotope area are a well-functioning, long-term contract program, which offers stability and pricing advantages; the resumption of calutron operations; the adoption of prorated conversion charges, which greatly improves the pricing of isotopes to small users; ISO 9002 registration of the IEF's quality management system; and a much more customer-oriented business philosophy. Efforts are also being made to restore and improve upon the extensive chemical and physical form processing capabilities that once existed in the enriched stable isotope program. Innovative ideas are being pursued in both technical and administrative areas to encourage the beneficial use of enriched stable isotopes and the development of related technologies.

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## 1. Introduction

A recent review of the history and background of the stable isotope program at the Oak Ridge National Laboratory (ORNL) was presented by Aaron et al. [1]. At that time, significant changes were taking place as a result of programmatic changes at the U.S. Department of Energy (DOE); unfortunately, most of those changes were having a negative impact on the availability of enriched stable and radioactive isotopes and related services. Significant changes are still continuing, but they have been of a more positive nature in the past couple of years. The changes involving enriched stable isotopes are in the calutron operations area, products and services capabilities, business practices, ISO 9002 registration, and new initiatives. While ORNL still produces and distributes radioisotopes, radioisotope operations at the Isotope Enrichment Facility (IEF) have been sharply reduced. However, the IEF maintains an important and active role in the radioisotope program since many radioisotopes are produced from an enriched stable isotope precursor.

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## 2. Calutron Operations

The calutrons currently being used for stable isotope enrichment, using the electromagnetic isotope separation (EMIS) process, were first placed in service in 1945 for uranium enrichment as part of the WWII Manhattan Project. This is the only production-level EMIS facility in the United States and one of only a few in the world. After a 3-year period of being in standby, the facility was restaffed and a 77-item readiness review for the resumption of calutron operations was conducted in late 1994. This formal readiness review examined operational capabilities, as well as environmental, safety, health, and quality issues, and had to be approved by both ORNL and the DOE. Calutron operations were resumed, on schedule, on January 3, 1995, with eight calutrons running for the separation of  $^{203}\text{Tl}$ , which is used in the production of  $^{201}\text{Tl}$  for cardiac imaging. Additional units were placed in operation, until a total of 16 calutrons were operating by June 1996. This represents 2 segments (8 calutrons per segment) in the segmented track, shown in Fig.1, which contains a total of 30 calutrons (3 sets of 8 calutrons and 1 set of 6 calutrons) and is an efficient and adequate operating level for the current circumstances.

In addition to receiving training to operate the calutrons, the largely new staff is performing a limited amount of development work to improve calutron operations. These developments include improved instrumentation and process monitoring capabilities, improved data management and interpretation, and the exploration of improved control systems. It should be pointed out that the level of product enrichment achieved following resumption exceeded expectations and past operational experiences.

A point has been reached where  $^{203}\text{Tl}$  inventory levels are adequate to permit the enrichment of other elements in support of the replenishment of general inventory levels and new market development efforts. Scheduling of new enrichment campaigns, both the elements to be run and the duration of the run, depends on a number of variables and is subject to change as customer needs change. A minimum campaign length of approximately 3 months is typical to justify the equipment changes that are needed for each element. It is possible to run different elements in the same segment as long as their masses are close enough to require the same magnetic field strength for separation.

## 3. Products and Services

Extensive chemical processing capabilities are an integral part of the isotope enrichment process since natural materials must be in the proper form to be placed into the calutrons for enrichment and the enriched isotopes must then be physically recovered from the calutron collector pockets and chemically purified to be placed into inventory. Enriched isotopes are generally inventoried in their most stable chemical form. Chemical processing is also required for most of the enriched isotopes that have been returned from customers using the DOE lease/loan programs. As a result of (a) the historical expertise developed in chemically processing so many elements and (b) experience with handling such valuable material in large quantities, the capabilities of the chemistry laboratory and staff gradually expanded to include many custom chemical preparations of enriched isotopes in chemical forms to meet customer

needs. When the calutrons were placed in standby in 1991, chemistry laboratory activities remained relatively stable because lease/loan returns, custom chemical preparations, and inventory processing work continued while the calutrons were in standby. At about the same time that the calutrons resumed operation in 1995 and required significant chemistry laboratory support, the level of lease/loan return work had decreased. This resulted in a relatively constant work load during the overall period and no significant staff size changes.

Similarly, extensive capabilities in chemical and physical form processing were developed over an approximately 35-year period by the Isotope Research Materials Laboratory (IRML) staff using metallurgical, ceramic, and high vacuum processing techniques. As a result of the reorganization of isotope activities at ORNL prompted by DOE's implementation of the Revolving Fund to finance Isotope Production and Distribution Program activities in 1990, the IRML was drastically reduced in size. As part of a consolidation effort to maintain capabilities but reduce costs, selected capabilities were relocated from several buildings at the main ORNL site to the IEF located at the Y-12 Plant site in Oak Ridge. Capabilities are being restored and, in many cases, enhanced as determined by resources and need.

The actual physical inventories of enriched stable isotopes (Sales and Research Materials Collection inventories) and the packaging and shipping functions were also relocated to the IEF from the main ORNL site. Locating the inventories, the chemical laboratories, and the materials laboratories all in the same facility has resulted in tremendous improvements in efficiency and was synergistic in improving capabilities. The three formerly separate organizations were also consolidated administratively into a Product and Services Group, offering enhanced flexibility in staffing and resources to better meet needs as they arise.

Among current initiatives of the Products and Services Group are the design and fabrication of a system for producing rare earth fluorides to permit the preparation of rare earth metals by Ca reduction of the fluorides [2]. In the past, La or Th was used as the reductant for most rare earth metals, but greater sensitivities regarding low-level radioactive contamination (both in the laboratory and the product) resulting from the use of Th has resulted in abandoning its use. Since La is not suitable to reduce all the rare earths, the fluoride conversion process was selected to permit high-efficiency, high-quality metal conversions of many of the rare earths. A new system for the reduction of Si, Ti, Zr, Hf, and possibly other materials, using the crystal bar reduction process [3], is also being designed and built.

The Product and Services Group was also responsible for dispensing actinide isotopes that had been enriched during past calutron operations. The actinide calutrons, which had been in stand-by since the early 1980s, were permanently shut down, and a large part of the inventory was moved to other facilities that have continuing actinide missions. This made it possible to classify the IEF as a "non-nuclear facility" and greatly reduced the level of resources that had to be expended on compliance activities (safety documentation, training, audits, etc.) without sacrificing the ability of ORNL to meet customer needs in the area of supplying enriched actinide isotopes. Cleanup of the actinide calutron area and glovebox laboratories is proceeding as resources are available.

#### 4. Business Issues

Like the materials processing groups, the ORNL Isotope Distribution Office (IDO), which handles all the business issues for the enriched stable and radioactive isotope distribution, also suffered significant cutbacks in 1990. As part of the consolidation effort to improve program efficiency, the IDO was also relocated from the main ORNL site to the IEF.

Shortly after the Revolving Fund was instituted, detailed pricing studies were undertaken, resulting in the development of a long-term contract concept. It was recognized that long-term contracts offered stability to operations and permitted planning to increase efficiency. It was also recognized that, as in most organizations with fixed and variable costs, the unit cost of production generally goes down as production increases, within the limits of production capability. The concept was implemented, and customers were offered long-term contracts that provided volume discounts for long-term commitments to buy large quantities of enriched isotopes. General guidelines for the volume discounts were developed based on committed production time measured in tank-hours (calutron operating hours), providing a common denominator that can be used for all isotopes, since each has a unique production rate in mass units. The long-term contract option has successfully been used by customers, both for their own consumption and for brokering of isotopes to other end users.

In another attempt to encourage the use of enriched isotopes, it was recognized that the effort and added expense to convert materials from their inventory form to an alternate form was, to some extent, independent of the size of the batch. In other words, the effort associated with the conversion of a 10-g batch was not significantly more than that for the conversion of a 10-mg or 100-mg batch. Also, the material loss rate for the conversion tended to go down as the batch size increased. It was decided that for alternate forms that had reasonable use rates and which could be stored without significant degradation, larger batches of material would be converted and maintained in inventory in the alternate form. The cost of the conversion and conversion losses would then be prorated over the entire batch, and the individual customer would only pay the prorated conversion and losses for the fraction of the larger batch that they purchased. For small users, this has resulted in orders-of-magnitude reductions in conversion costs and loss charges. For larger users, the conversion costs are no more than they would have been for the large-scale conversion. Delivery times for selected alternate forms have also improved since they can often be shipped from existing inventory instead of being scheduled for custom conversion.

Another area of improvement that is in progress involves information management for both inventories and business operations. ORNL has over 2000 individual batches of enriched isotopes in inventory that have been produced over the operating history of the IEF. While detailed information or "pedigrees" of these batches are generally available, they were not always easily and quickly retrievable. All batches are being reviewed, and more comprehensive information is being maintained in a readily accessible manner. The ORNL IDO is sponsoring this activity and incorporating it in an overall business information system that will automate and integrate virtually all information from an initial customer contact through shipping and billing of a product.

## **5. ISO 9002 Registration**

One of the most far-reaching initiatives to be undertaken in the program for stable isotope enrichment and distribution at ORNL since the resumption of calutron operations was a decision to seek ISO 9002 (International Organization for Standardization) registration of its quality assurance system. The ISO 9002 registration is awarded by an accredited third party, in this case, Underwriters Laboratories Inc., after thorough and rigorous auditing of the quality management system and its implementation against an internationally accepted standard [4]. Periodic audits, every 6 months, verify continued functioning of the quality management system and are required to maintain registration. Once the decision to seek registration was made, a year-long effort of evaluating every aspect of stable isotope enrichment and distribution was undertaken. In most cases, it was found that formalization of existing practices was all that was required to meet this standard. In other cases, significant development of new mechanisms and quality system components was required. It is a commonly held conclusion that going through this process improved the organization, in addition to achieving customer satisfaction by preventing nonconformity at all stages from production through distribution.

## **6. Summary**

While past financial problems encountered by the ORNL enriched stable isotope production and distribution program resulted in placing the calutrons in standby and caused significant reductions in both capabilities and personnel, it also permitted a reinvention of the program after more favorable conditions developed. As a result of operational and business reviews and the opportunity to rebuild the program, significant improvements have been achieved and are continuing. The consolidation of all activities in one facility, the execution of long term contracts with volume discounts, the continuing restoration of selected capabilities, and an emphasis on efficiency, flexibility, quality, and customer satisfaction have placed this program in an excellent position to meet the future needs of enriched stable isotope users in research, medicine, and industry around the world.

## **Acknowledgement**

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## **References**

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- [2] F. H. Spedding and A. H. Daane, The Rare Earths (John Wiley, New York and London, 1961), 78-87, 102-112.
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Figure 1. View of the calutron track used for stable isotope enrichment. The four separate segments of the track, divided by magnetic shunts (under the sets of steps in the center of the track), are visible.

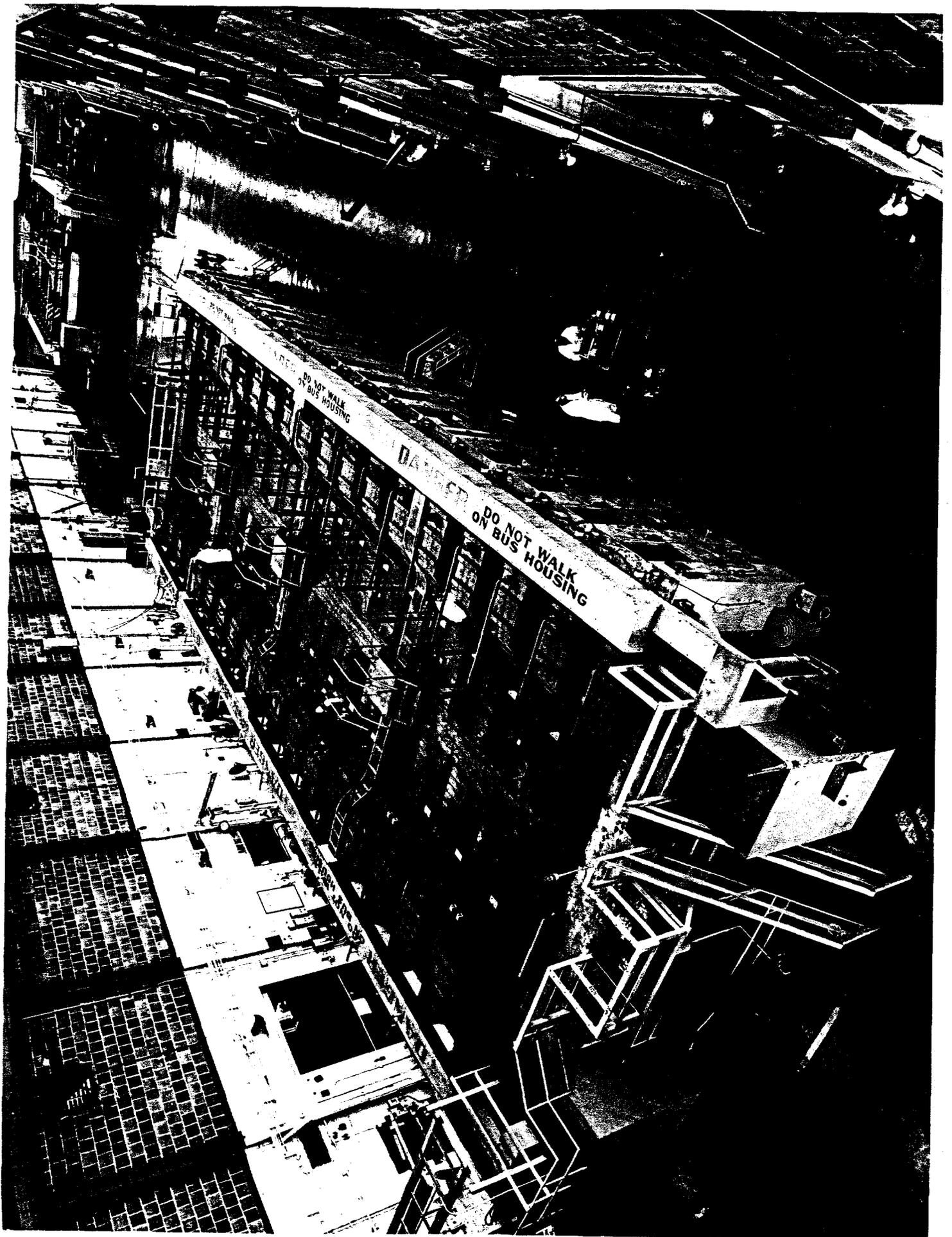


Fig. 1