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# STABLE ISOTOPE ENRICHMENT – CURRENT AND FUTURE POTENTIAL

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## ABSTRACT

Oak Ridge National Laboratory (ORNL) operates the Isotope Enrichment Facility for the purp of providing enriched stable isotopes, selected radioactive isotopes (including the actinides), isotope-related materials and services for use in various research applications. ORNL is responsi for isotope enrichment and the distribution of approximately 225 nongaseous stable isotopes from multi-isotopic elements. Many enriched isotope products are of prime importance in the fabricat of nuclear targets and the subsequent production of special radionuclides. State-of-the-art techniq to achieve special isotopic, chemical, and physical requirements are performed at ORNL. This rep describes the status and capabilities of the Isotope Enrichment Facility and the Isotope Resea Materials Laboratory as well as emphasizing potential advancements in enrichment capabilities.

## DISCLAIMER

## 1. Introduction

Enriched stable isotopes, isotope related materials, and services provided by the Oak Ridg National Laboratory (ORNL) under the direction of the U.S. Department of Energy's (DOE) Offic of Isotope Production and Distribution (OIPD) are crucial to many areas of research, medicine, an industry. Nuclear targets using stable isotopes are vital to many branches of nuclear researc including physics, chemistry, material science, medicine, biology, geoscience, and engineering. Thes targets are also useful in the production of radioactive isotopes, which are used predominately i health-care delivery programs. Timely availability of enriched stable isotopes is essential, and th capability of ORNL to continue to provide the materials necessary to meet the changing needs of th research, medical, and industrial community is a subject of concern. The purpose of this paper is t review ORNL's Stable Isotope Enrichment and Distribution Program in regard to its ability to mee current and future needs of the user community. To that end, this discussion will include: (1) th current status of the Isotope Enrichment Facility (IEF) and Isotope Research Materials Laborato (IRML) and (2) potential advancements in enrichment capabilities.

The objective of ORNL's Isotope Enrichment Program (IEP) is to enrich and distribute som 225 stable isotopes from approximately 50 multi-isotopic elements. These objectives are achieve through utilization of a portion (~5%) of the Electromagnetic Isotope Separation (EMIS capabilities, which were constructed as part of the wartime effort during the early 1940's. While few isotopes are used in large quantities and have dedicated enrichment processes to meet volum needs, the EMIS process is particularly amenable to providing relatively small quantities of a broa spectrum of highly enriched stable isotopes to meet the needs of a wide range of basic and applie research and commercial applications. The EMIS process is applicable to all elements in the periodi table. All isotopes of each multi-isotopic element can be enriched simultaneously. This process is further characterized by very high separation factors in a single pass. Other physical and chemical

enrichment methods provide larger throughput, but require multistaging to obtain high enrichments and are either isotope selective or alter the feed abundance to enhance either the heaviest or lightest isotope.

## 2. Electromagnetic Isotope Separation

EMIS is a well-known process, and detailed descriptions have been published in numerous articles in the past. The intent here is to provide only a brief review for the reader who is unfamiliar with this separation process.

The EMIS process is based on the motion of an energetic ion in a magnetic field. The radius of curvature, $r$, for a particle of mass, $m$, and charge $e$ moving in a magnetic field of strength, $B$, with a velocity, $v$, is given by the relationship $mv^2/r = Bev$. Charged particles are formed in an ion source and accelerated in an electrostatic field that is generated by potential difference, $V$, resulting in an energy, $E$, for all isotopes as given by $E = Ve = 0.5\ mv^2$. From these two equations, we can determine the radius of curvature to be $r = (2\ mV)^{0.5}/Be^{0.5}$. Thus, the change in radius for each isotope is given as $\Delta r \propto \sqrt{\Delta m}$ .

Primary components of an electromagnetic isotope separator are an ion source, an electrostatic accelerating system, a magnetic field, a vacuum system, and a series of collector pockets. Most production-type isotope separators employ a side-extraction plasma discharge ion source with a slit aperture that produces a line image at the collector. The plasma is maintained by electrons emitted from a hot cathode, typically tantalum or tungsten, in a magnetic field in the direction of the electron flow. To maintain copious ion formation and a stable discharge, feed materials and a vapor transport system must be chosen to provide a pressure in the plasma region of $\sim 10^{-3}$ torr. The ionized particles are extracted from the ion source and accelerated to their final energy in a geometry that produces a line image near the extraction slit. After being deflected by the analyzing magnetic

field, the individual isotopic species are refocused at the collector defining slits. The ion beams are intercepted by collector pockets constructed of either carbon, copper, or aluminum. These collectors may require cooling to dissipate the heat produced by ion bombardment.

Electromagnetic isotope separators require very precise magnetic field adjustments to cause the beams to focus at the collector. This effect is obtained by well-defined iron shims usually placed inside the vacuum chamber. Shims can be designed for: (1) multibeam separation, (2) both radial and axial focusing, or (3) the source and collector to be external or internal to the analyzing magnet. Separators with the source and collector internal to the magnetic field have a higher degree of beam neutralization and shorter beam paths, which minimize losses due to scattering and charge exchange (and, in turn, contribute a higher beam intensity at the collector).

### 3. Enrichment Capabilities

The original objective of the IEP at ORNL was to provide enriched stable isotopes to government research institutions. Later ORNL's mission included providing enriched stable isotopes, selected radioisotopes (primarily the actinides), and isotope related materials and services for world-wide distribution for research, medicine, and industrial applications. To accomplish this task, the IEP at ORNL uses a portion of the electromagnetic isotope capabilities that was built in the 1940s at the Oak Ridge Y-12 Plant for the enrichment of uranium-235. The Y-12 Plant, at its peak in 1945, had 1152 operational high-current mass spectrometers, known as calutrons (*Cal*ifornia *U*niversity Cyclo*trons*), housed in nine production buildings. The production calutrons were of two types: (1) the 122-cm-radius alpha calutron used for initial enrichment, and (2) the 61-cm-radius beta machine used for final enrichment. The current IEP uses a building that contained 72 of the beta-type separators.

The original facility layout consisted of two magnetic tracks, each containing 36 calutrons in two 30.5-meter-long parallel arrays. The two sides were magnetically coupled by a 9-meter-long magnetic yoke to complete the magnetic circuit. This arrangement was not amenable to multi-element separation requirements and two major modifications were made. The first modification included altering the magnetic circuits on one track for stable isotope enrichments and part of the second track for selected radioactive actinide isotopes enrichments. Concurrent with these efforts, support facilities, such as shops and chemistry laboratories for processing calutron feed materials and products for both stable and actinide enrichments, were constructed.

Multi-element enrichments of stable isotopes are obtained by subdividing one track into four segments by removing six separators and installing three magnetic shunts from one side of the track to the other. Three of the segments contain eight calutrons each, and one segment contains six separators. The second track is subdivided into three segments. One of the segments is housed in the containment area and has been used for selected radioactive isotope separations, primarily the actinides. The other 2 segments, containing a total of 24 separators (some in varying states of disrepair) are available for special experiments and separations. As a result of these two modifications, the current facility has the capability for meeting the requirements of separating stable isotopes and selected radioactive isotopes. ORNL has the only known facility outside the former Soviet Union (FSU) with such capabilities.

The IEF contains three types of isotope separators. These include 32 machines of the 180° homogeneous magnetic field-type, 6 separators with 255° inhomogeneous magnetic field, and 1 sector-type separator. The 180° calutron has a beam radius of 61 cm, uses a side-extraction ion source, and produces a line image at the collector. This machine has a 180° homogeneous magnetic field and employs linear shims to produce magnetic field aberrations at the 90° area of the beam, which provides focus and allows for two arc capabilities. With the source located internally with the

analyzing magnet for maximum beam neutralization, beam currents of >100 mA can be achieved at the collector. The second type separator that is routinely used for enrichments is the 255° inhomogeneous magnetic field separator, which has an inhomogeneity of 0.5 where the magnetic field is defined by $B_z = B_0(r_0/r)^n$, with the subscript *0* referring to the reference orbit and *n* referring to the inhomogeneity. The six separators of this design have both radial and axial focusing at the collector as well as a greater mass dispersion and resolution, and they are employed when high isotopic enrichments are of prime importance. The sector separator has an inhomogeneity of 0.8, a beam resolution of approximately ten times that of the standard calutron at about one tenth of the beam current. This machine with its source and collector external to the analyzing magnet, has the capacity to either postaccelerate or decelerate the ions at the collector, which makes it useful for production of ion-implanted or surface-deposited targets.

The temperature range of the original calutron source has been expanded to operate from ambient temperature to greater than 1300 K. Proper feed selection and *in situ* halogenation techniques in conjunction with the expanded temperature range is sufficient to form volatile feed for all the multi-isotopic elements in the periodic chart. High process efficiency ion sources and sources designed to process milligram quantities of feed have been developed. This development, coupled with efficient (>90%) feed recovery techniques, make high-enrichment, dual-pass separations, and separations involving rare and expensive feed materials feasible.

Comprehensive chemical processing capabilities are an integral part of the isotope enrichment process. Over the years, extensive , specialized processing capabilities have been developed at the IEF to support enrichment operations and to meet the special chemical form needs of the customers. Chemical processing performed at the IEF includes: (1) calutron feed material preparation, (2) recovery and purification of enriched isotopes from the calutron receiver pockets, (3) recovery

and purification of loaned isotopes upon their return from uses, and (4) chemical conversions to provide isotopes in noninventory chemical forms as needed by a customer.

Complementary (and in a few cases, duplicate) processing capabilities focusing on physical rather than chemical form processing were developed at the X-10 Site and have been performed at IRML. A wide variety of vacuum, metallurgical and ceramic processing methods were developed and applied to both enriched and natural abundance stable and radioactive isotopes. Among the custom order materials preparations that have been supplied are: vacuum processed and rolled thin films for use as nuclear targets; metal melting, casting and working of custom shapes and compositions; hot pressing or sintering of special ceramic forms and compositions; in-core neutron dosimeters; and application of equipment and expertise to a wide variety of basic and applied research projects at ORNL and private laboratories. Strong research and development (R&D) programmatic support from DOE (from several materials-related sources) was instrumental in developing and maintaining these diverse capabilities.

## 4. Isotope Enrichment Program: Background, and Current Status

The IEP was initiated as a totally funded program sponsored by the Atomic Energy Commission (AEC) — the predecessor to the DOE. Emphasis was on R&D of EMIS technology as well as advancement of new and improved products to be made available to government research institutions. As production increased, demands for these new materials expanded, and isotopes were sold to nongovernment, domestic research institutions with revenues returned to the U.S. Treasury. A loan program was established to make materials available for nondestructive experiments for DOE or other institutions where the research was of interest to DOE. Subsequently, sales distribution was expanded to include industrial institutions and foreign users, and the revenues were returned to the program to offset production cost. Prices for isotopes were established to recover the full cost of

production at the time the enrichment was performed. DOE funding was provided to support R&D activities, cover capital purchases and facility upgrading, and underwrite the production, loan, and distribution program.

Current financial support for all government isotope activities in the United States is provided by a revolving fund that was established by the U. S. Congress in 1989. All costs incurred, including capital purchases, facility upgrades, R&D, and compliance issues, as well as direct production costs are supported by this fund. Revenues from all isotope materials and services are returned to the revolving fund. Naturally, under these conditions, emphasis is placed on the production of products that have commercial applications and produce high revenue.

At the time the revolving fund was instituted, DOE consolidated all its isotope activities under one program, the Isotope Production and Distribution Program (IPDP), and founded OIPD to provide oversight responsibilities for these activities. OIPD reviewed all of its operations with regard to capability to produce sufficient revenue to offset operating cost, age, and condition of facilities, and liability associated with continued operation. To maintain a viable revolving fund, major changes were initiated that involved realigning and consolidating a number of activities, terminating those activities that were determined to be a liability, and establishing new pricing policies for stable isotopes and isotope related materials and services. At ORNL, a number of radioactive isotope operations were curtailed or moved to other DOE laboratories, IRML activities were severely reduced, the Isotope Distribution Office (IDO) was consolidated in the IEF for efficiency, and the IEF continued enrichments at a reduced level. New pricing policies were established, and the loan program was replaced by a lease arrangement.

The revolving fund that was established as a one time appropriation from Congress has encountered problems in maintaining a balance between costs and revenues generated from sales of products. This difficulty is prompted by reduction in sales due to price increases, the influx of

isotopes through joint ventures with the FSU, and increased operating expenses resulting from spiraling costs associated with environmental, health, and safety concerns. For the fund to remain solvent, further reductions in ORNL's isotope activities have been necessary. These include placing the calutron operations in standby as of September 1991, with a consequent transfer of personnel and loss of technical expertise, suspension of IRML activities, and curtailment of further consolidation efforts. The status of stable isotope activities at ORNL is summarized in Table 1.

## 5. Future Potential

Recent events in the scientific community, political arena, and the commercial sector have effected the isotope program at ORNL. In February 1991, a workshop was convened at the National Academy of Science to discuss the availability of isotopically enriched materials. The workshop participants proposed a study by the National Research Council to look at the broad, long-term issues concerning production of enriched isotopes in the United States over the next few decades. One of this study's goals would be the careful evaluation of the option that involves the future of the calutrons. On August 12, 1992, a Congressional subcommittee hearing was convened to discuss DOE's isotopes program. The committee addressed all isotope programs, both stable isotope enrichment and radioisotope production and identified the problems associated with total dependance on the revolving fund for maintaining a viable program to meet the nation's needs. More recently, DOE/ORNL has consummated a multimillion dollar, long-term contract for providing stable isotopes to a commercial sector customer.

To meet the near-term program objectives, plans are being formulated to restart the calutrons some time in mid-1993. Efforts for consolidation have been reinitiated, and most of the IRML equipment used for stable isotopes has been moved to the IEF. This equipment will be installed and

activated as needed and funded to meet customer demands. Restaffing of both technical and support personnel is underway, and training programs are being prepared.

Future advancements in isotope enrichment could be accomplished at ORNL by utilizing the Plasma Separation Process (PSP) equipment currently stored at the K-25 Site in Oak Ridge. Exploiting this technology, in conjunction with the calutrons, would provide new and better products at reduced prices, make available the isotopes necessary to meet to changing needs of the national and international community, and increase the isotope program's customer base. This equipment is currently being moved to the IEF at the Y-12 Plant, and except for the 45-ton cryogenic magnet, the move should be completed in November 1992.

The PSP is based on the ion cyclotron resonance of a charged particle in a uniform magnetic field. The ion cyclotron resonance frequency is given by the relationship: $\omega = CB/M$, where $\omega$ is the gyration frequency, $C$ is a constant, $B$ is the magnetic field strength, and $M$ is the mass of the particle. If an electric field is applied at a fixed gyration frequency, the magnetic field can be varied to cause the particle of interest to resonate and absorb energy. (Fig. 1).

The primary components of the plasma separation system consist of: the source, containing the feed plate and the microwave antenna; the excitation region, with a uniform magnetic field and resonant electric field; and the collector made up of product and tail plates. The solenoidal magnetic field is produced by a pool-boiling, cyrostable, nonpersistent, superconducting magnetic that is 8 meters long with a clear bore of 1 meter. The resonant electric field is produced by helical-wound drive coils. Detailed descriptions of the process are given in other papers and will not be discussed here.

The process was developed and tested by TRW, Inc., Los Angeles, CA under contract from DOE as part of the Advanced Isotope Separation (AIS) Program to develop and test alternative methods for enriching uranium at lower cost. The PSP proved successful in demonstrating scientific

feasibility using the preprototype hardware and a production prototype module (PPM) was designed and built. Ultimately, the Atomic Vapor Laser Isotope Separation (AVLIS) process was chosen for development for future uranium enrichment, and the PSP equipment was made available for other applications.

The PSP is isotope selective and is particularly adaptable to the enrichment of the metallic elements. Flexibility of the PPM was demonstrated by TRW in production enrichments of nickel-62, and palladium-102 in 9 months with low upfront costs. Enrichment of isotopes of many other elements have been demonstrated or modeled using the PPM. (Fig. 2). By comparison, the enrichment factor for the PSP is approximately one tenth that of the calutron, but it has throughput capabilities of approximately 1,000 production calutrons. This high production rate could augment the present enrichment program, making available large quantities of material at low to moderate enrichments and providing pre-enriched feed material for the calutrons. Pre-enriched feed for the calutrons would greatly increase the cost effectiveness of the high enrichment.

Assembly and operation of the PSP equipment is contingent on identifying large scale isotope needs and obtaining commitment from the potential users to obtain program support. Four such uses have been identified, and meetings with potential users are planned to discuss quantities, cost production schedules, etc. If it is determined feasible, the machine would be assembled in the IEF to use existing support facilities, such as chemistry laboratories, shops, and utilities, and to take advantage of the existing staff that could be shared between the calutron and the PSP operations. The conceptual design for the installation of the PSP in the IEF is shown. (Fig. 3). Funding proposals for the installation, renovation, and startup of this process have been prepared and submitted.

## 6. Conclusion

This paper has attempted to provide a brief overview of the IEP at ORNL in regard to its current technical capabilities and status as well as potential enrichment capabilities. Current policies mandate that all isotopes, isotope products, and services recover full replacement cost of the operation. The program possesses the technology to increase efficiency through consolidation of efforts and use of the PSP to provide new and better products at lower cost. The future of the program will depend on the commitment of Congress and DOE in addition to the support of the user community.

TABLE 1
ISOTOPE ENRICHMENT FACILITY, STATUS

| | |
|---|---|
| **Calutron Separations** | |
| Stable Isotope | Standby |
| Actinide | Shutdown |
| **Chemical Processing** | |
| Stable Isotope | Active |
| Actinide | Shutdown |
| **Technical Services** | |
| Chemical Services | Active |
| IRML | Semi-active |
| **Product Distribution (IDO)** | Active |

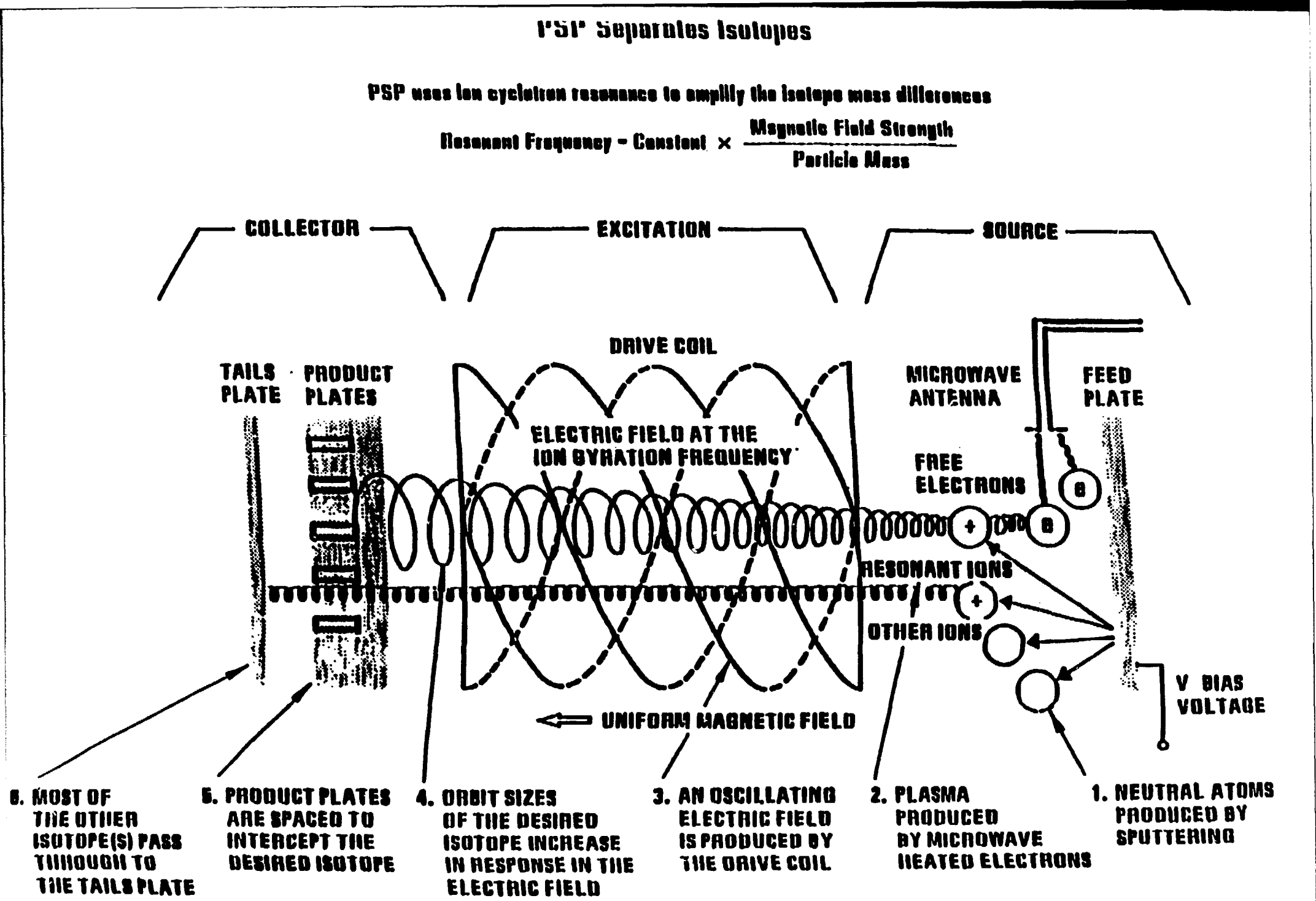
"SI" Separates Isotopes
PSP uses ion cyclotron resonance to amplify the isotope mass differences
Resonant Frequency = Constant × Magnetic Field Strength / Particle Mass
COLLECTOR
EXCITATION
SOURCE
DRIVE COIL
TAILS PLATE
PRODUCT PLATES
MICROWAVE ANTENNA
FEED PLATE
ELECTRIC FIELD AT THE ION GYRATION FREQUENCY
FREE ELECTRONS
RESONANT IONS
OTHER IONS
V BIAS VOLTAGE
UNIFORM MAGNETIC FIELD
6. MOST OF THE OTHER ISOTOPE(S) PASS THROUGH TO THE TAILS PLATE
5. PRODUCT PLATES ARE SPACED TO INTERCEPT THE DESIRED ISOTOPE
4. ORBIT SIZES OF THE DESIRED ISOTOPE INCREASE IN RESPONSE IN THE ELECTRIC FIELD
3. AN OSCILLATING ELECTRIC FIELD IS PRODUCED BY THE DRIVE COIL
2. PLASMA PRODUCED BY MICROWAVE HEATED ELECTRONS
1. NEUTRAL ATOMS PRODUCED BY SPUTTERING

Fig. 1

# PSP Has Separated Over 20% of the Elements on the Periodic Table

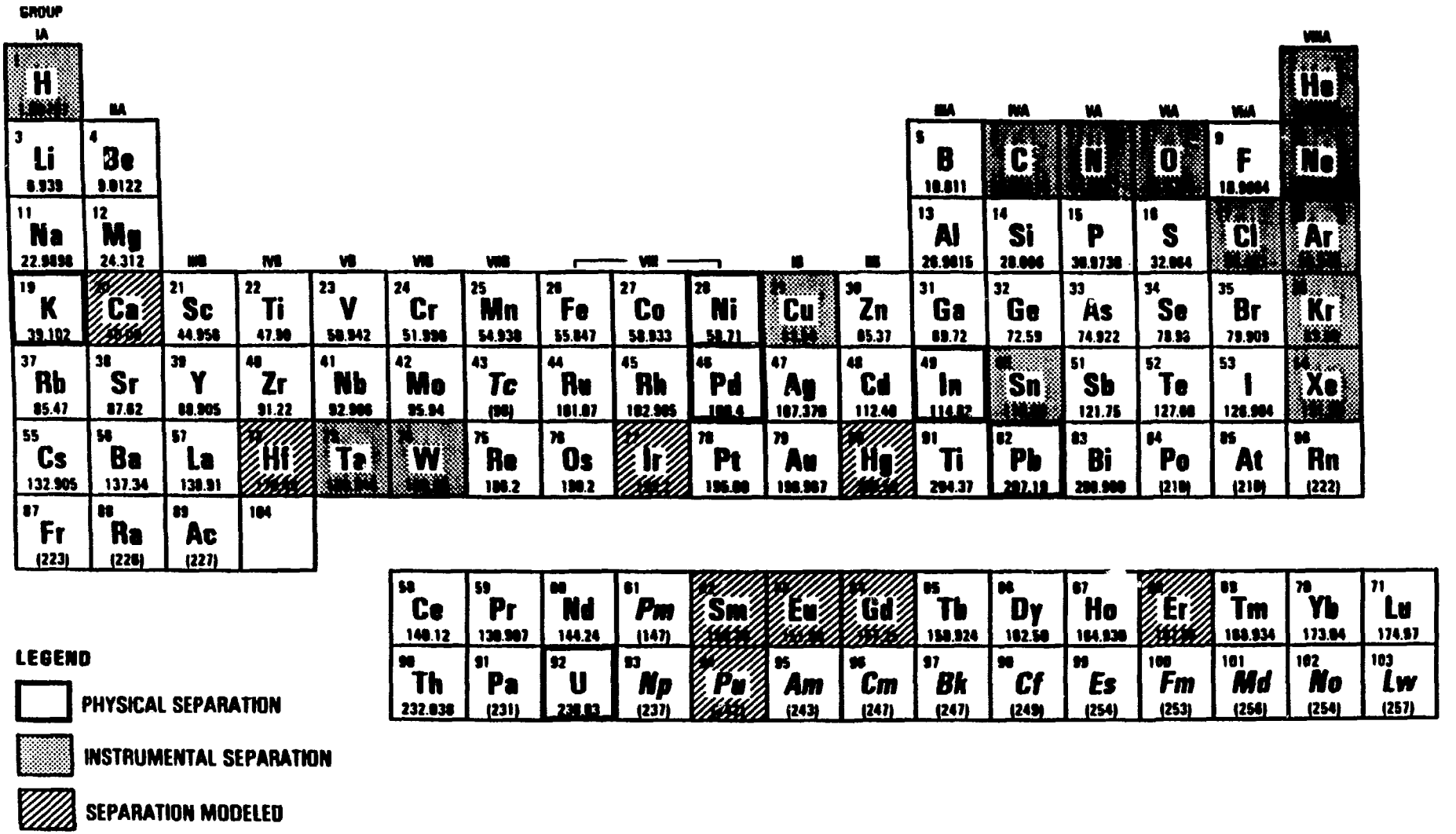


Fig. 2

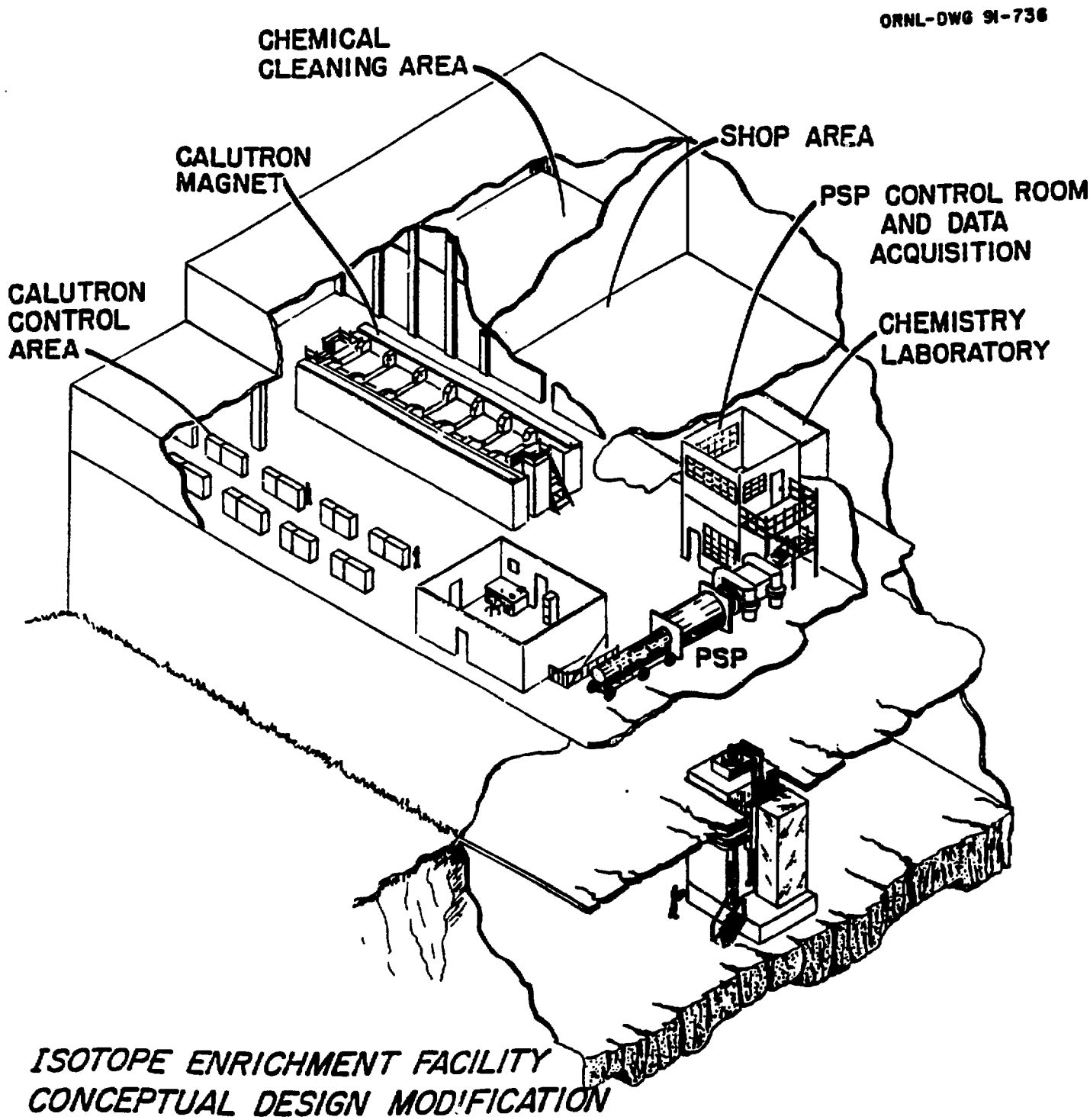
ORNL-DWG 91-736
CHEMICAL CLEANING AREA
SHOP AREA
CALUTRON MAGNET
PSP CONTROL ROOM AND DATA ACQUISITION
CALUTRON CONTROL AREA
CHEMISTRY LABORATORY
PSP
ISOTOPE ENRICHMENT FACILITY
CONCEPTUAL DESIGN MODIFICATION

Fig. 3