TARGETS PRODUCED IN MASS SEPARATORS*

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MASTER

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Summary

Isotopically enriched nuclear targets are prepared directly in the electromagnetic mass separators by allowing the resolved ion species to impinge on a suitable substrate. The calutron, a multi-milliamper mass separator, has been successful in producing ion-implanted targets for nuclear research and in the doping of semiconductors. By using flat magnetic shims, a rotating target holder, and a sliding indexing shutter, multiple targets with uniform implantation have been achieved. The typical accelerating energy for this method is 40 keV, although this can be increased by using multiply-charged ions. For highly enriched targets from materials where the desired isotope is of low natural abundance, the 180° Oak Ridge Sector Isotope Separator has proven particularly successful. This machine, which is an exceptionally high-resolution separator, is interfaced with an on-line computer for surveillance and control. Isotonic enhancement factors of 4×10^5 have been achieved for targets prepared in this machine. Targets are prepared by placing the host material (typically carbon, aluminum or nickel foils) at the receiver, 1 cm above the normal straight-line beam path. The desired ion species is steered onto the target through an electrostatic beam deflection system, thereby minimizing neutral particle contamination. The energy of an ion at impact, and hence the deposition depth, is controlled by applying a suitable potential (-40 kV to +40 kV) at the collector. Ion deposition energies from a few eV to several hundred keV have been achieved. High-energy implants (>80 keV) are made by post-acceleration of a multiply-

charged ion beam of the specified isotope. Energies of 350 keV have been obtained for ^{22}Ne by collecting the +5 ion with a total accelerating voltage (source plus collector potential) of 70 keV. A typical target made in the separator has a line shape which measures 1.5 mm x 1 cm on the substrate, a thickness of 20 to 100 $\mu\text{g}/\text{cm}^2$, and an isotopic purity >99%. The deposition line can be as long as 8 cm and the width can be altered, within limits, by the aperture dimensions and the target placement relative to the mass analysis slit system.

Introduction

The wide range of basic and applied scientific research, in conjunction with the various types of acceleration systems, has led to an increase in the number and variety of nuclear targets produced and used in recent years. The diversity of target requirements along with the widely differing chemical and physical properties of both the target and host material has precluded any single method for the preparation of all targets. The technology exists however, using a number of techniques, to produce targets of every element in the periodic chart that will meet most user requirements. One method used at the Oak Ridge National Laboratory and many other facilities throughout the world, is to prepare isotopically enriched targets directly in an Electromagnetic Separator.

The isotope separation facility (Fig. 1) at Oak Ridge uses three types of electromagnetic isotope separators, the calutrons, 255° double focussing machines and the 180° Oak Ridge Sector Isotope Separator (ORSIS). These separators, in contrast to most laboratory machines, are designed and built for higher throughput and are used primarily for providing isotopically enriched samples for basic research and as precursors for nuclear medicine.

Fig 1

However, a limited amount of effort is directed to target preparation which has proven successful in producing both ion implanted and thin film nuclear targets.

Target Preparation in Separators

Targets are prepared in the mass separator by placing the host material at the collector and directing the desired ion species onto the backing by adjusting the accelerating voltage and magnetic field. While this is the basic premise of the preparation, it in itself is not sufficient to produce targets that would have a wide application to scientific research. Through the use of special techniques and certain modifications to these machines, target thickness can be increased, uniformity assured, and higher chemical and isotopic purity attained.

Targets may be prepared in mass separators by two different methods. These are, ion implantation and surface deposition. The difference is that ion implantation is carried out at higher energies (keV) and the ions are trapped in a thin layer just below the surface while the surface deposition method uses low energies (eV) and the material is deposited on the surface of the substrate. In either case, the host material must be electrically conductive since the deposition is made with charged particles.

For ion implantated targets, the ions strike a surface which is usually placed perpendicularly to the direction of the ion. Each impinging ion striking the surface undergoes a slowing down by a series of collisions and is either trapped in the substrate lattice or backscattered after one or more collisions. The distribution of the implanted ion is approximately Gaussian in shape and the mean depth is determined by the ion energy and the mass and atomic numbers of the ion and substrate atoms. At the beginning of

the bombardment the trapping efficiency is approximately 100% for most commonly used host materials. As the run progresses, the collection efficiency decreases and after a sufficiently long bombardment time, saturation of the substrate is reached. Saturation is reached when for every impinging ion a target atom must leave. Trapped atoms leave the substrate by diffusion or by release with sputtered atoms. Saturation, sputtering phenomenon, and ion stopping in solids have been studied by many authors and are described in detail in the literature.

The thickness of targets made by the ion implantation technique is limited by the saturation value for the particular target and host material. At ion energies of 50 keV (50 kV is assumed to be an approximate accelerating potential for many isotope separators) a thickness of only a few $\mu\text{g}/\text{cm}^2$ can be achieved. This value, unfortunately, is too thin for many experiments. Since the penetration depth and hence the amount of material collected before saturation is proportional to the ion energy, ion implanted targets are often made at higher energies. This is accomplished by collecting an ion species with a higher charge state or postaccelerating the resolved ion beam at the collector or, a combination of both.

Surface depositions have no theoretical limitations on thickness but in practice, careful consideration must be given the physical strength of the host material.

Thin film or surface depositions are prepared by applying a positive potential to the collector to reduce the ion energy at impact. This retarding potential is typically a few tens of volts less than the accelerating potential. The volatility and the self sputtering ratio are used as the criteria for determining the ion energy.

Several years ago, the major demand for ion implants was for the doping of semiconductor materials with only a moderate interest in ion implanted targets. This work was carried out in the calutron and provided the basic techniques for our ion implantation work.

The calutron is a 180° homogeneous magnetic-field separator which utilizes a side-extraction slit-type ion source and produces a line image at the collector. The source and receiver are in the main magnetic field (Fig. 2). This arrangement eliminates problems of secondary electron emission at the collector. The separator accelerates singly charged particles to energies up to 40 keV with controllable beams of on the order of 100 mA.

Special collectors which continuously move the targets are used for most implantations. This movement along with collimators to limit the angular acceptance, result in a uniform deposition over large areas.

As a result of the commercial development of ion-implant machines and the increasing need for thicker surface deposited targets with high isotopic enrichment, a separator with different characteristics than the standard calutron is required.

Target Preparation in the 180° Sector

Of the three types of separators at ORNL, the one that best fulfills the current needs of target-making is the 180° Sector Separator (ORSIS). This machine, shown schematically in Fig. 3, is mounted in the gap of a beta-type calutron magnet and uses an inhomogeneous magnetic field with a field inhomogeneity, n , of 0.8. That is;

$$B_z = B_o \left(\frac{r_o}{r} \right)^n$$

where the subscript o refers to reference orbit. In brief, the separator has

ORNL-DWG 73-5698

Fig 2

SCHEMATIC OF THE 180° ORSIS

Fig 3

an orbit radius of 61 cm, a 180° deflection angle, an object-to-image distance of ~700 cm, and an acceleration voltage of 40 kV. The ion source for the separator is of the electron bombardment type and employs side-extraction slit geometry. Beam currents at the receiver are typically on the order of a few milliamperes, which is about one-tenth the output of the isotope production calutron separators for the same element. The mass dispersion of the separator is equal to $427 \left(\frac{\Delta m}{m} \right)$ cm and can resolve the masses to give enhancement factors up to 4×10^5 . Enhancement factors as defined here involve the final assay and initial abundance in the following way:

$$\text{Enhancement Factor} = \frac{C_f (100 - C_o)}{C_o (100 - C_f)},$$

where C_o and C_f are the initial and final abundances of the isotope. A scan showing the separation and resolution of three tin isotopes is presented in Fig. 4.

This separator system, with its modifications, utilizes three high-voltage supplies. One supply provides control to the source unit, while the other two supply postacceleration potential, deaccelerating potential, beam deflection voltage, and sweep voltage. The particular configuration used is determined by the type of target needed.

Retardation potential is supplied to the collector assembly using a nominal 40 kV supply identical with the accelerating voltage supply. This configuration, as opposed to the more straightforward method of biasing the accelerating voltage to the collector, is more difficult and expensive but offers an advantage which will be discussed later. Ion energies are accurately measured by using an auto-ranging differential digital volt meter.

Fig. 4

The receiver assembly, which was constructed specifically for target preparation work, is designed such that the target and collector box (which houses the targets) are mounted on insulators capable of withstanding a potential of >40 kV. Figure 5 shows a schematic of the receiver arrangement. Other features of the receiver include: (1) negative electrodes, (2) ground electrodes, (3) shutter, and (4) monitoring probes. These elements perform the following functions, respectively: (1) suppress electrons which would cause erroneous readings, and steer and sweep the ion beam; (2) isolate the electric field from the ion beam; (3) cover and protect the target when conditions are not optimized; and (4) maintain the beam in the proper position.

A PDP-8/I computer with memory, storage, and peripheral equipment is interfaced with the separator for surveillance and nominal control. The control functions extend from the ion source to the control of the ion energy at impact, which is particularly important when preparing highly enriched isotopic surface-deposited targets. The operating system applies to any collection by providing the necessary information concerning the particular target specifications, such as the ion energy, the desired nuclide, and the maximum and minimum ion currents required.

In preparing a particular target, charge materials of high chemical purity are chosen in an effort to minimize contamination due to complex ion formation or by isobars which appear in the same mass spectrum. Source and collector parts are thoroughly cleaned to remove any residual material from previous collections. The target holder is vacuum outgassed to prevent charge exchange, which results in false ion current readings.

The target backings are mounted on a holder and placed in the collector box (See Figs. 6 and 7) and is further outgassed by allowing an inert gas beam, usually argon, to impinge on the holder 1 cm below the target area. When

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FIG. 4 Exploded View of Target Holder

FIG. 8 ORSIS Target Holder

outgassing is complete the magnetic field and accelerating voltage are set for the desired ion species and the computer determines when all conditions are within prescribed limits and then opens the shutter. The beam is steered onto the target area by adjusting the deflection voltage. The number of ions striking the target(s), and hence the quantity of the deposit, is constantly monitored by using a current integrator.

Surface depositions and low-energy (below 40 keV) ion-implanted targets are prepared by applying a positive retarding potential to the collector. In preparing targets for surface deposition, special emphasis is placed on accurately determining and controlling the ion energy. Ion energies are constantly monitored throughout the preparation and periodic calibration checks are made to compensate for any drift resulting from temperature changes.

In the event that medium-energy ions (40 to 80 keV) are required to meet the user's specifications, these are obtained by reversing the potential at the receiver so that the particles are postaccelerated into the particular substrate. For energies above 80 keV, it is necessary to select a multiply-charged ion of the desired isotope. In order to increase the production of multiply-charged ions, arc power supplies are available to increase the electron energy in the arc up to 600 eV.

An inherent benefit of retardation is that it acts as an energy barrier and is effective in reducing isotopic contamination caused from ion scattering during transmission and from ions formed in the accelerating region having less than the nominal accelerating energy. Further, both chemical and isotopic contamination arising from neutral vapor, charge exchange, and sputtering from the internal parts of the separator can be minimized by deflecting the resolved ion beam from its normal straight line path.

Finally contamination caused from high voltage discharge at the source is brought under control by the use of a separate power supply for deacceleration potential. Ions formed during source sparking have less than normal energies. However, since the retardation voltage is constant during this time, ions cannot penetrate the potential if;

$$V_r > V_a - \frac{\Delta M}{M} V_a$$

where V_a and V_r are the acceleration and deacceleration voltages, respectively, M the mass of the target ion and ΔM the mass difference between the target ion and the next heavier ion. While we have made no attempt to determine these benefits individually, an example of their collective effects is given in Table 1.

A number of thin, surface-deposition targets have been prepared in the ORSIS in the last several years, primarily for coulomb excitation studies. These targets typically have a line shape which measures 1.5 mm x 1 cm. They are deposited on suitable backing materials which are 20 to 100 $\mu\text{g}/\text{cm}^2$ thick. Depositions of 25 to 75 $\mu\text{g}/\text{cm}^2$ of the desired isotope with ion energies <200 eV are typical of the thin targets prepared in the separator. Targets with these specific ions will be used as examples in the following discussions.

Random mass assays of microgram-sized samples have been obtained from target slits mounted inside the decel receiver box and directly in front of the target. Typical assay results are shown in Table 2. It should be noted that the ion beam did not strike directly all the surfaces where the samples were taken; thus it seems reasonable to assume that the isotopic purity of the target itself would be slightly higher than shown here.

SUMMARY OF COLLECTED ^{156}Dy

	Ion Energy (eV)	156 %	158	160	161	162	163	164	Total enrichment
180° ORSIS	325	99.534	0.0228	0.0656	0.1768	0.0882	0.0550	0.058	407,000
Decel Receiver	650	99.26	0.12	0.10	0.23	0.16	0.08	0.06	256,000
	325	98.692	0.0356	0.135	0.464	0.262	0.206	0.205	144,000
	325	95.867	0.055	0.306	3.230	0.354	0.117	0.070	44,000
Regular Receiver	40,000	74.2	0.2	1.6	6.3	6.4	5.4	5.8	5,500
Calutron									
2nd Pass		93.2	0.04	0.25	2.0	1.6	1.4	1.5	220
2nd Pass Charge		5.8	0.30	3.0	16.0	25.9	24.5	24.5	
1st Pass		20.9	0.58	3.43	19.70	22.20	16.90	16.30	500
Natural Abundance		0.0524	0.0902	2.294	18.88	25.53	24.97	28.18	

Table 1

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TABLE 2

Assay Comparison of ORSIS Targets
and Calutron Separated Isotopes

Isotope	Natural Abundance Charge (%)	ORSIS Target (%)	Calutron Single Pass (%)	Calutron Second Pass (%)
^{156}Dy	0.0524	99.534	20.9	93.2
^{158}Dy	0.0902	98.91	23.78	~95.0 *
^{162}Er	0.136	99.26	20.4	98.96
^{164}Er	1.56	99.56	62.7	99.77
^{154}Gd	2.15	99.36	66.95	99.35

* Estimated

Every effort is made to avoid chemical contamination of targets. To date, we know of only two instances where impurities were present. In each case, the source of these impurities was traced to the charge material. The charge contained small quantities of chemical contaminants which formed molecular ions having the same mass as the target isotope. A special effort is made to ensure that this will not recur.

The amount of material deposited on the substrate is determined by the use of a current integrator. Deposited samples, needed for other reasons, have been chemically recovered and analyzed by isotopic dilution methods. The quantity recovered from decel-type collections has been within 10% of the monitored values. While we can verify the amount collected, we have not attempted to measure the uniformity of the deposit; however, it is known that the beam intensity over the area is homogeneous, which should result in a reasonably uniform layer. The deposition area is clearly outlined by defining slits placed ~ 1 mm in front of the target. Figure 8 shows a typical surface deposition target on a $60\text{-}\mu\text{g}/\text{cm}^2$ carbon backing. Many other surface depositions have been made with a variety of specifications, such as size, shape, ion energy, thickness, and backing material. The target shape and size can be altered by changing the defining slits, by moving the target behind the focal point, or by applying a sweep voltage to the electrode in the collector region. Ion energies as low as 50 eV and deposits as thick as $1\text{ mg}/\text{cm}^2$ have been achieved in reproducible experiments.

In addition to targets prepared by the surface deposition of enriched isotopic material, ion-implanted targets ranging from a few keV to 350 keV have been made. For example, Ne^{5+} ions were collected using a total accelerating voltage of 70 kV. The maximum ion energy that can be obtained is the product of the total accelerating voltage and the charge state. The particle current, which

FIG. 8 Typical Surface Deposition Target

is highly dependent on the charge state, differs considerably from element to element.

The effort required in making targets in the separator by direct deposition varies considerably from target to target and is dependent on a number of factors. Some of the most important of these are: natural abundance of the desired isotope, ion output of the element involved, number of targets which can be prepared either simultaneously or with the same source and receiver assembly, and the amount of material deposited.

One of the most attractive features of ion-deposited targets, especially those prepared by deacceleration, is the high isotopic purity of the deposited material. As indicated in Table 1 & 2, the same (or higher) assay quality can be obtained directly in the Sector as in two passes through an isotope production separator. Target preparation in the separator is a full-cost recovery operation that must integrate man-hours involved in equipment assembly and actual separator operation. The cost for preparing targets is usually more expensive than by other methods and the problems of transporting thin foils has limited their use to some degree.

Conclusion

The ORSIS is important to the nuclear research community because of its capability for supplying highly enriched targets (>99%) in a single preparation starting with a feed having a very low (<1%) natural abundance. In addition, the thin, uniform deposits can be highly attractive in the field of heavy-ion research.