TARGET SELECTION FOR THE HRIBF PROJECT*

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ABSTRACT

Experiments are in progress at the Oak Ridge National Laboratory (ORNL) which are
designed to select the most appropriate target materials for generating particular
radioactive ion beams for the Holifield Radioactive Ion Beam Facility (HRIBF). The 25-
MV tandem accelerator is used to implant stable complements of interesting radioactive
elements into refractory targets mounted in a high-temperature FEBIAD ion source
which is on-line at the UNISOR facility. These experiments permit selection of the target
material most appropriate for the rapid release of the element of interest, as well as

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realistic estimates of the efficiency of the FEBIAD source. From diffusion release data information on the release times and diffusion coefficients can be derived. Diffusion coefficients for Cl implanted into and diffused from CeS and Zr$_5$Si$_3$ and As, Br, and Se implanted into and diffused from Zr$_5$Ge$_3$ have been derived from the resulting intensity versus time profiles.

INTRODUCTION

The Holifield Radioactive Ion Beam Facility (HRIBF), now under construction at ORNL [1], will have the capability of generating short-lived radioactive nuclei by irradiation of targets with H$^+$, D$^+$, 3H$^+$, and 4He$^+$ ion beams from the Oak Ridge Isochronous Cyclotron (ORIC). The targets are close-coupled to an ion source which will ionize the radioactive species upon diffusion from the target material and transport to the ionization chamber of the source. Experiments have been initiated which utilize the HHIRF tandem accelerator to implant stable complements of interesting radioactive atoms into refractory targets mounted in the UNISOR [2] version of the FEBIAD [3] ion source. Experiments similar to those performed at GSI [4] have been carried out; they are designed to measure the time evolution and ionization efficiencies of implanted species diffused from refractory target materials which are initial candidates for forming radioactive beams at the HRIBF. These experiments allow selection of the refractory target material most appropriate for swift release of the element of interest, as well as realistic estimates of the efficiency of the FEBIAD source for providing ion beams of the species prior to actual radioactive beam generation in the HRIBF. A fraction of the implanted species, which continuously diffuse from the target, are ionized, accelerated, and mass analyzed. The signals derived from the experiments are used to determine the time release profiles. The total release time of the species from the target material involves both diffusion from the solid and hold-up times between the target and the
ionization chamber of the source due to surface adsorption processes. However, the method described in this report is only valid whenever the diffusion time $\tau_d$ is much longer than the effusion delay time $\tau_e$ and the transit time $\tau_t$ from the ion source to the detection system or $\tau_e$ and $\tau_t$ are known.

Data derived from ion implantation experiments has been analyzed and used to extract diffusion coefficients by use of a computer code which solves Fick's second law (see, e.g., Ref. 5) as represented by the one-dimensional form of the following equation:

$$\frac{\partial n}{\partial t} = D \frac{\partial^2 n}{\partial x^2} - \lambda n$$

where $n$ is the concentration and $\lambda n$ accounts for losses when the atom is radioactive. Diffusion coefficients of ion implanted Cl in CeS and Zr$_5$Si$_3$ and As, Br, and Se in Zr$_5$Ge$_3$ were determined. The results were then used to model the release of radioactive ion beams from homogeneously distributed planar and spherical geometry targets which represent the actual distributions and geometries which will be used for RIB generation at the HRIBF.

EXPERIMENTAL APPARATUS AND EQUIPMENT

The 25-MV tandem accelerator at the HHIRF is utilized to implant stable complements of interesting radioactive atoms into refractory targets maintained in the FEBIAD ion source which is the "on-line" ion source used at the UNISOR facility. The experiments described in this paper were performed by implanting Cl$^{8+}$ into CeS and Zr$_5$Si$_3$ and As$^{13+}$, Br$^{13+}$, and Se$^{13+}$ ions into Zr$_5$Ge$_3$ targets maintained in the anode structure of the ion source. The ion implanted beams pass through a thin window of a high Z
material before entering the target, which slightly degrades the energy of the ion beam. The thickness of the window, usually tantalum, is typically 3.5 to 5 mg/cm². The final energy after passing through the window is chosen so that the ions penetrate to a depth of 15 to 35 μm in a thin circular disk (~9 mm in diameter and ~1.5 mm in thickness) of the material of interest. The depth of particle penetration within the target material simulates the average diffusion distance that an atom or molecule would have to travel before exiting a spherical macro-particle of the same radius. The simulation program TRIM [6] is used to determine the position and distribution of the implanted species within the target material.

Upon entry into the target, the neutral species diffuse rapidly in the high temperature sample (typically 1645°C). Beam heating effects raise the target temperature in proportion to the beam power; typically a 10 Watt beam raises the temperature by an additional 50°C. The target thus resides at ~1700°C during the implantation. Approximately 50% of the particles reach the entry side of the target and are evaporated and channeled into the anode region of the ion source where they are bombarded with an electron beam accelerated from a hot cathode. A fraction of the particles are ionized and accelerated from the ion source and mass analyzed. Data concerning the time-release of the diffusing species is thereby gathered and analyzed. The magnitude of the time dependent mass analyzed signal is used to determine the time-release profile of the particular species.

RESULTS

A simulation program called DIFFUSE [7] which solves Fick's second equation was used to calculate release curves for the indicated species diffusing from CeS, Zr₅Ge₃, or Zr₅Si₃. The implantation distribution function was determined from the final position
of particles as computed from the TRIM code. The input parameters required by DIFFUSE include the particle depth and distribution within the target, the activation energy, and the temperature of the sample. In order to determine the diffusion coefficient \( D \), an initial value is assumed; this value is then iteratively altered until an acceptable fit to the experimental data over the full diffusion range is found.

Targets that have been bombarded for extended periods of time were found to have larger diffusion coefficients than those for new targets. This effect is believed to be the consequence of the presence of impurities in new targets which compete for vacancies created by the ion beam. After prolonged bombardment, these impurities are driven out of the diffusing medium, thereby increasing the number of vacancy sites which enhances the diffusion of the implanted species. For targets subjected to several minutes of ion bombardment, the release profile could be replicated by fits with a single diffusion coefficient. Figure 1 shows a typical time release profile for \(^{78}\)Se diffusing from a \( \text{Zr}_5\text{Ge}_3 \) target; the solid lines represent fits to the experimental data. The fits provide good to excellent agreement with the corresponding experimental data. From fits to these data, diffusion coefficients \( D \) were derived. The diffusion coefficients, ranges of implantation, and release times for these projectile/target combinations are listed in Table I. The release time \( \tau \) is defined as the time required to release 50% of the steady-state ion beam intensity diffused from the target [8]. The diffusion coefficients derived here are similar in magnitude to those found in the literature for other species/refractory target combinations.

The distribution function for ion implanted targets is sharply Gaussian and very discretely localized in position beneath the target surface. During operation of the HRIBF, the radioactive species will be uniformly distributed within either planar or spherical targets. Therefore, the release times will differ from those obtained from ion
implantation experiments. Thus, the diffusion coefficients derived from the ion implantation experiments are extremely valuable for estimating the release times of uniformly distributed radioactive species in planar and spherical targets. Comparisons of numerical simulations for various geometries are shown in Fig. 2. It is obvious that macro-particle target materials are desirable for the release of radioactive elements if temperatures can be kept low enough to avoid sintering.

CONCLUSIONS

The results of the present experiments clearly demonstrate that the ion implantation technique can be utilized as a practical and cost-effective means for evaluating candidate refractory targets for releasing specific elemental materials prior to their actual use in generating radioactive in beams. Diffusion coefficients extracted from these data can also be used to optimize the target particle size and geometry in order to minimize the release time of the element of interest from the candidate target material.

REFERENCES


2. UNISOR, Oak Ridge Associated Universities, Oak Ridge, Tennessee 37831, USA.


7. TRIM - the Transport of Ions in Matter, J. F. Ziegler, IBM Research, Yorktown Heights, New York 10598, USA.

8. Diffuse is a program that solves one- and three-dimensional forms of Fick's second equation. The code was written by G. D. Alton, J. Dellwo, and I. Y. Lee.


FIGURE CAPTIONS

Fig. 1. Typical time-release profile of $^{78}$Se diffusing from Zr$_5$Ge$_3$. The theoretical fits to the experimental data were determined by solution of Fick's second equation using the program DIFFUSE [6]. $\circ$: experiment; $-$: theoretical; implantation depth: 17.9 $\mu$m; target temperature: 1675°C; diffusion coefficient: $D = 5.1 \times 10^{-8}$ cm$^2$/s.
Fig. 2. Theoretical time-release spectra for homogeneously distributed $^{78}\text{Se}$ and $^{71}\text{Se}$ diffusing from 20-mm and 200-mm-thick planar and spherical geometry $\text{Zr}_5\text{Ge}_3$ targets as predicted by the program DIFFUSE [6]. Target temperature: 1675°C; Diffusion coefficient: $D = 5.1 \times 10^{-8}$. 
Table 1. Times $\tau$ and Diffusion Coefficients $D$ for Selected Ion Implanted, Candidate RIB Species/Target Material Combinations

<table>
<thead>
<tr>
<th>Species</th>
<th>Target Material</th>
<th>Implantation Depth ((\mu)m)</th>
<th>Target Temperature ($^\circ$C)</th>
<th>$D$ (cm$^2$/s)</th>
<th>$\tau$ (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cl</td>
<td>Zr$_5$Si$_3$</td>
<td>31.3</td>
<td>1695</td>
<td>$1.2 \times 10^{-5}$</td>
<td>13</td>
</tr>
<tr>
<td>Cl</td>
<td>CeS</td>
<td>2.6</td>
<td>1690</td>
<td>$4.4 \times 10^{-10}$</td>
<td>170</td>
</tr>
<tr>
<td>As</td>
<td>Zr$_5$Ge$_3$</td>
<td>18</td>
<td>1670</td>
<td>$5.1 \times 10^{-8}$</td>
<td>39</td>
</tr>
<tr>
<td>Br</td>
<td>Zr$_5$Ge$_3$</td>
<td>16</td>
<td>1675</td>
<td>$5.2 \times 10^{-7}$</td>
<td>7</td>
</tr>
<tr>
<td>Se</td>
<td>Zr$_5$Ge$_3$</td>
<td>18</td>
<td>1670</td>
<td>$5.1 \times 10^{-8}$</td>
<td>40</td>
</tr>
</tbody>
</table>
Implantation Depth: 17.9 \mu m; \quad D=5.1 \times 10^{-8} \text{ cm}^2/\text{s}

![Graph showing relative intensity over time with experimental data and theoretical fit.]

**Figure 1**
Time Release Profiles for Selected Stable and Radioactive Isotopes of Se

- **0.02mm sphere**
- **0.02mm disk**
- **0.2mm sphere**
- **0.2mm disk**

- **76Se, \( \tau_{1/2}=\text{stable} \)**
- **75Se, \( \tau_{1/2}=4.74\text{m} \)**

**Figure 2**
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