Computational Design of High Efficiency Release Targets for Use at ISOL Facilities

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This report describes efforts made at the Oak Ridge National Laboratory to design high-efficiency-release targets that simultaneously incorporate the short diffusion lengths, high permeabilities, controllable temperatures, and heat removal properties required for the generation of useful radioactive ion beam (RIB) intensities for nuclear physics and astrophysics research using the isotope separation on-line (ISOL) technique. Short diffusion lengths are achieved either by using thin fibrous target materials or by coating thin layers of selected target material onto low-density carbon fibers such as reticulated vitreous carbon fiber (RVCF) or carbon-bonded-carbon-fiber (CBCF) to form highly permeable composite target matrices. Computational studies which simulate the generation and removal of primary beam deposited heat from target materials have been conducted to optimize the design of target/heat-sink systems for generating RIBs. The results derived from diffusion release-rate simulation studies for selected targets and thermal analyses of temperature distributions within a prototype target/heat-sink system subjected to primary ion beam irradiation will be presented in this report.

INTRODUCTION

Many of the reactions, fundamentally important in nuclear physics and astrophysics, are inaccessible to experimental study using stable/stable beam/target combinations and, therefore, can only be studied with accelerated radioactive ion beams (RIBs). The availability of RIBs offers unique opportunities to further our knowledge about the structure of the nucleus, the stellar processes that power the universe, and the nucleosynthesis burn-cycles responsible for heavy element formation. The well-known on-line isotope separator (ISOL) technique, used for the generation of short-lived radioactive nuclei, involves a multi-step process: the nuclear reaction products must diffuse to the surface of the target material, evaporate from the surface, and effusively flow to the ion source where a fraction of the species of interest are ionized, extracted from the source and mass analyzed prior to post acceleration. The principal means whereby short-lived radioactive species are lost between initial formation and utilization are associated with the diffusion and surface adsorption processes where the hold-up times are long with respect to the life-time of the species of interest. Both diffusion and surface adsorption processes depend exponentially on the operational temperature of the target/ion source, and thus, successful RIB generation requires careful consideration of the physical, chemical, and metallurgical properties of the target material and thoughtful design of highly permeable target matrix systems with short diffusion lengths and controllable target temperatures. In this report, we briefly review our efforts to develop targets that simultaneously

incorporate these properties so that useful RIB intensities for a wide range of short-lived species can be generated for use in the astrophysics and nuclear structure physics research programs at the Holifield Radioactive Ion Beam Facility (HRIBF).

HIGHLY PERMEABLE TARGET MATRICES

Efforts are underway at the HRIBF to design fast release targets with controllable temperatures for optimum production and generation of RIBs [1]. The speed of release is dependent on the diffusion constant, the target temperature, and the thickness through which the radioactive particles must pass prior to arriving at the surface of the target material. Therefore, fast release times are synonymous with large diffusion constants, short diffusion lengths (i.e., small target material dimensions), and low enthalpies of adsorption for released products.

Thin Al_2O_3 fibrous targets have been successfully used during on-line production and release of ^{17}F and ^{18}F for use in the HRIBF astrophysics research program [2]. Other refractory fibrous metal oxides, such as ZrO_2 , HfO_2 are also being considered for use as target matrices for the production of F isotopes. Still, other fibrous targets such as SiO_2 , TiO_2 , TaO_3 , Y_2O_3 as well as rare-earth can be used. Many other refractory materials such as the metal-carbides are under consideration for RIB applications. We have also chosen low-density carbon fiber, such as carbon-bonded-carbon fiber (CBCF) or reticulated vitreous carbon

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fiber (RVCF), to serve as the target material plating matrix and thermal transport conduit for heat removal. These fibrous materials are refractory and highly porous (density typically 0.06 to 0.12g/cm³). Very thin layers of selected target materials, for example, Ni, SiC and UC₂, have been uniformly deposited onto RVCF and CBCF to form composite target matrices using either electroplating (EP) or chemical vapor deposition (CVD) methods [3].

DIFFUSION RELEASE

Diffusion in an isotropic medium is governed by Fick's laws. For the release of radioactive isotopes of decay constant λ , the time-dependent diffusion equation, modified to include the rate of production S and the rate of decay loss E , can be expressed in a Cartesian coordinate representation as follows:

$$\frac{\partial n}{\partial t} = D \left(\frac{\partial^2 n}{\partial x^2} + \frac{\partial^2 n}{\partial y^2} + \frac{\partial^2 n}{\partial z^2} \right) + S(x, y, z, t) - E(x, y, z, t) \quad (1)$$

where n is the concentration of the diffusing substance; D is the diffusion coefficient, assumed to be independent of concentration. For an uniform distribution of radioactive species produced with a primary ion beam of intensity I and charge Z , the production rate S is given by

$$S(x, t) = \frac{\sigma n I L}{ZeV} \quad (2)$$

where e is the charge on the electron, n is the density of interaction nuclei, I is the intensity of the primary ion beam, L is the length of the target material, σ is the cross section for production of the species of interest, and V is the volume over which the species are distributed. The decay loss E is identically equal to

$$E(x, t) = n\lambda \quad \text{where } \lambda = 0.693/\tau_{1/2} \quad (3)$$

for radioactive species with half-life $\tau_{1/2}$.

The rate of diffusion release from a target material can be calculated by solving the appropriate form of Eq. 1. Diffusion in composite RVCF targets coated with selected target materials can be modeled by solving the one-dimensional equation for a planar geometry target. For an initially homogeneous distribution of nuclei in an infinite planar target of thickness l , the fractional release $f(t)$ is given by

$$f(t) = 1 - \frac{2}{l^2} \sum_{n=0}^{\infty} \frac{\exp\{-\mu_n Dt\}}{\mu_n} \quad (4)$$

where $\mu_n = (n+1/2)^2 \pi^2 / l^2$.

We have assumed that the species do not diffuse into the carbon substrate but are reflected at the boundary and that surface desorption is fast compared to the mean diffusion time. The total release rate per unit surface area can be written in the form

$$R(t) = \frac{2S}{l} \sum_{n=0}^{\infty} \frac{1 - \exp\left\{-\left(\mu_n + \frac{\lambda}{D}\right)Dt\right\}}{\mu_n + \frac{\lambda}{D}} \quad (5)$$

The fractional release $f(t)$ of nuclei uniformly distributed within a fibrous cylindrical geometry metal-oxide target material of radius r , can be represented by the summation

$$f(t) = 1 - \frac{4}{r^2} \sum_{n=1}^{\infty} \frac{\exp\{-\alpha_n^2 Dt\}}{\alpha_n^2} \quad (6)$$

where α_n s are the positive roots of Bessel functions of the first kind of zero order, $J_0(\alpha_n r) = 0$. The total release rate per unit length on the fiber material then becomes

$$R(t) = 4\pi S \sum_{n=1}^{\infty} \frac{1 - \exp\left\{-\left(\alpha_n^2 + \frac{\lambda}{D}\right)Dt\right\}}{\alpha_n^2 + \frac{\lambda}{D}} \quad (7)$$

Figure 1 shows the fractional release, $f(t)$, of Cu from RVCF coated, respectively, with 1 μm and 2 μm of Ni. The fractional release of fluorine from a 6 μm diameter Al₂O₃ fibrous target is shown in Fig. 2. Estimated total diffusion release rates from a number of targets are given in Table 1. Although the diffusion release rates from these highly permeable targets have been calculated in the frame-work of a simple diffusion model, the results show that the diffusion release of short-lived radioactive species such as ⁵⁸Cu and ¹⁷F from these target matrices are sufficiently fast to meet the intensity requirements for research programs at the HRIBF.

Fig.1. Fractional release of Cu from a composite RVCF matrix coated with 1 μm and 2 μm Ni, respectively. Diffusion coefficient $D = 1.53 \times 10^{-9} \text{ cm}^2/\text{s}$.

Fig. 2 Fractional release of fluorine from 6 μm diameter fibrous Al_2O_3 target material. Diffusion coefficient: $D = 10^{-10} \text{ cm}^2/\text{s}$.

and equilibrium temperature distributions in target/heat-sink systems.

A prototype target/heat-sink system now under design at the HRIBF is displayed in Fig. 3. The device can be inserted into or removed from the target/ion source through a vacuum inter-lock port. The primary beam, which produces the radioactive species, passes through a thin window and deposits energy in the target material during the slowing down process. The length of the target material is chosen so that the beam exits the target with ~ 4 MeV energy and comes to rest in a C-beam stop that is connected to a Cu heat-sink. The target material reservoir can be resistively heated when necessary to maintain the desired target operational temperatures.

Computational studies have been conducted to simulate the heat transport and temperature distributions in several targets designed for use at the HRIBF. These targets include: fibrous Al_2O_3 for ^{17}F production using low-energy

TABLE 1. Estimated diffusion release rate of selected radioactive species from fibrous and composite targets.

Element	Target	Ion Beam	Prod. Rate/ μA	Release Rate/ μA	Temp ($^{\circ}\text{C}$)	D(cm^2/s)
^{17}F	Al_2O_3 Fiber	40 MeV Deuterons	1.53×10^{10}	2.9×10^9	1430	10^{-11}
^{18}F	Al_2O_3 Fiber	40 MeV Protons	1.46×10^{10}	1.3×10^{10}	1430	10^{-11}
^{58}Cu	2 μm Ni / RVCF	30 MeV Protons	5.5×10^8	2.1×10^8	1300	1.529×10^{-9}
^{132}Sn	15 μm UC_2 / RVCF	60 MeV Protons	4.2×10^8	2.8×10^8	2000	8×10^{-8}
^{132}Sn	15 μm UC_2 / RVCF	1 GeV Protons	4.0×10^9	2.6×10^9	2000	8×10^{-8}

TARGET/HEAT-SINK SYSTEM

Higher temperatures significantly decrease diffusion release times of particles from targets. Thus, in order to optimize radioactive species release, it is desirable to operate the target to the temperature limit set by the vapor pressure of the target material which does not compromise the ion source efficiency. The limiting vapor pressure of the CERN-type electron-beam-plasma ion source, presently used at the HRIBF, is 2×10^{-4} Torr [4]. It is also important to provide means for removal of beam deposited heat from the target matrix at controlled rates so that, as high as practical, primary beam intensities can be used to produce the species of interest. Several computer codes have been used to simulate heat generation, heat removal, and to estimate equilibrium temperature distributions in target matrices. Heat deposited in the target material by the primary ion beam is simulated by use of the Monte Carlo code, Stopping and Range of Ions in Matter (SRIM) [5]. The heat generation function, derived from SRIM, takes into account the axial energy deposition, dE/dx ; effects due to the radial profile of the primary ion beam; and the scattering of the primary ion beam during passage through the target which results in broadening of the beam. The finite element code ANSYS [6] is used to simulate heat transfer, temperature gradients

deuteron beams; Ni coated RVCF for ^{58}Cu production using low energy protons; and UC_2 coated RVCF composite targets for production of fission products with 1 GeV protons. The maximum target temperature versus beam power in a 2.4 cm long fibrous Al_2O_3 target irradiated by 20 MeV deuterons, as calculated with ANSYS, is displayed in Fig. 4. The energy of the ion beam was chosen for the production of ^{17}F isotopes via the nuclear reaction $^{16}\text{O}(d,n)^{17}\text{F}$. The operating temperature of the target is set by the vapor pressure limitation of the ion source system (~ 1650 $^{\circ}\text{C}$). As shown in Fig. 4, this temperature is already reached with less than 45 W of deuteron beam power, limiting the primary deuteron beam intensities that can be used for the production of ^{17}F . This is due to the fact that these highly permeable fibrous materials have low thermal conductivities. Partially coating the Al_2O_3 matrix with a thin layer of Ir can be an effective means for removal of heat from the target. As also shown in Fig. 4, the maximum beam power can be increased to 150W by adding a 25 μm thick Ir foil spiral rolled between 1.5 mm thickness layers of the Al_2O_3 fibrous matrix. An interesting result, derived from the analysis, is that radiative heat transfer is more effective than conduction even at temperatures less than 1600 $^{\circ}\text{C}$ for low thermal conductivity fibrous target materials such as Al_2O_3 .

Fig. 3. Schematic drawing of a removable/insertable composite target/heat-sink system showing the components which make up the system now under design at the HRIBF for use in RIB generation. The target material may be particulate, fibrous, or plated onto composite target support disks.

The beam heating results in a non-uniform temperature distribution profile, both radially and axially. It is desirable to smooth out the temperature distribution in the target material. One approach for achieving this effect is to coat the entrance end of the beam-stop with pyrolytic carbon with the high thermal conductivity direction oriented parallel to the face of the beam-stop. Comparisons of the radial and longitudinal temperature profiles with and without pyrolytic carbon show that pyrolytic carbon is effective in smoothing out the temperature distribution in target systems such as $\text{Al}_2\text{O}_3/\text{Ir}/\text{RVCF}$ and $\text{UC}_2/\text{Ir}/\text{RCVF}$.

Fig. 4. Maximum target temperature versus beam power in a 2.4 cm long fibrous Al_2O_3 target spiral rolled with 25 μm thick Ir and without Ir, irradiated by 20 MeV deuterons.

ACKNOWLEDGEMENTS

Research at the Oak Ridge National Laboratory is supported by the U.S. Department of Energy under contract DE-AC05-96OR22464 with Lockheed Martin Energy Research Corp.

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