ADVANCED TARGET CONCEPTS FOR RIB GENERATION RECEIVED

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In this report, we describe highly permeable composite target matrices that simultaneously incorporate the short diffusion lengths, high permeabilities, and heat removal properties necessary to effect maximum diffusion release rates of short-lived, radioactive species as required for efficient radioactive ion beam (RIB) generation in nuclear physics and astrophysics research programs. The RIB species are generated by either fusion or fission nuclear reactions between high energy ^1H , ^2H , ^3He or ^4He ion beams and specific nuclei which make up the target material. The target materials may be used directly as small diameter particulates coated or uncoated with Re or Ir to minimize adsorption following diffusion release and eliminate sintering of the particulates at elevated temperatures; plated onto both sides of thin disks of C, for example; or plated, in thin layers, onto low density, Ir or Re coated carbon-bonded-carbon-fiber (CBCF) or reticulated-carbon-fiber (RCF) to form sponge-like composite target matrices; or in other cases, where applicable, the target material of interest can be grown in crystalline fibrous form and fabricated in woven mats of the target material to form a highly permeable fibrous structure.

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 which power the universe and the nucleosynthesis burn cycles responsible for heavy element formation. As a consequence of world-wide interest in the potential benefits of using RIBs, facilities have been built, funded for construction or proposed for construction in Asia, Europe, and North America [1]; these include the Holifield Radioactive Ion Beam Facility at the Oak Ridge National Laboratory (ORNL) [2]. These facilities are based on the use of the well-known on-line isotope separator (ISOL) technique in which radioactive nuclei are produced by nuclear reactions in selectively chosen target materials by high-energy proton, deuteron, He, heavy ion beams or neutrons. The ISOL technique has been utilized to produce low energy ion beams from over 600 isotopes [3]. In order to successfully perform nuclear physics and astrophysics research with radioactive ion beams (RIBs), intensities ranging from 10^5 to 10^{12} particles/s must be delivered to the experimental station. While these intensities appear modest, they are not easily achievable because of the limited production rates of the species and their lifetimes in relation to the times required for diffusion from solid state targets and effusion into the ion source. The production rates in the target are set by practical limits on the primary beam intensity in terms of the maximum permissible, on-target, power density

which can be used without compromising the efficiency of the ion source or the physical integrity of the target, and the reaction cross sections for producing the species of interest. The radioactive product beam intensity is determined by the rate at which the particular species can be diffused from the target, the average residence time of the radioactive species on surfaces between the target and the ionization chamber of the source in relation to their lifetimes, the ionization efficiency of the ion source, and losses in beam transport between the ion source and experimental station. The upper temperature at which the target/ion source can be operated without deleteriously affecting the ionization efficiency is set by the vapor pressure characteristics of the target material [4, 5]. Because of these factors, careful attention must be given in the selection of the most appropriate target material and in the design of composite target/heat-sink systems which incorporate the short diffusion lengths and high permeability attributes required for prompt and efficient diffusion release of the species of interest at controlled temperatures.

In this report, we briefly review present efforts at the HRIBF to design advanced highly permeable composite targets required for effecting fast and efficient diffusion release of the radioactive species of interest from the target material.

PROCESSES WHICH AFFECT RELEASE TIMES

The principal means whereby short half-life radioactive species are lost between initial formation and utilization are associated with diffusion and surface adsorption processes where the delay times are long with respect to the life-time of the species in question for forming RIBs. The diffusion and surface

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adsorption/desorption processes depend exponentially on the operational temperature of the target/ion source. Because of the strong temperature dependence of the adsorption process, the materials of construction of the target chamber, vapor transport tube and ionization chamber of the source can significantly affect the residence times of the radioactive species through the magnitudes of the enthalpies of adsorption on the surfaces of materials that the particle comes in contact with following diffusion release and transport to the ionization chamber of the source. The diffusion and surface adsorption processes are briefly described below.

Diffusion Theory

The time and temperature-dependent release of a nuclear reaction product species, embedded in a chemically dissimilar target material, implies the presence of a binary diffusion mechanism which underlies the release process. Whenever there is a concentration gradient of impurity atoms or vacancies in a solid material, the atoms or vacancies will move through the solid until equilibrium is reached. The net flux, J , of either the atoms or the vacancies is related to the gradient of concentration, ∇n , by Fick's first equation given by:

$$J = -D\nabla n \quad (1)$$

where D is the diffusion coefficient.

The time-dependent form of Eq. 1 is known as Fick's second equation. The three-dimensional form of this equation, which allows for the creation of particles $S(x,y,z,t)$ as well as the loss of particles $E(x,y,z,t)$, can be expressed in a Cartesian coordinate representation as follows:

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

where D is assumed to be independent of concentration.

The target geometries that will be used or are envisioned for use at the HRIBF include: planar, cylindrical and spherical geometries. For example, the time-dependent form of Eq. 2 appropriate for planar targets is given by

$$\frac{\partial n}{\partial t} = D \frac{\partial^2 n}{\partial x^2} + S(x,t) - E(x,t) \quad (3)$$

The equation, appropriate for diffusion from cylindrical geometry targets, where the diffusion process is assumed to move only in the r direction, can be expressed in the following form

$$\frac{\partial n}{\partial t} + \frac{D}{r} \frac{\partial}{\partial r} \left(r \frac{\partial n}{\partial r} \right) + S(r,t) - E(r,t) \quad (4)$$

While for spherical-geometry targets, where the diffusion process is assumed also to be solely in the radial direction, Fick's second equation takes the form:

$$\frac{\partial n}{\partial t} + \frac{E}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial n}{\partial r} \right) + S(r,t) - E(r,t) \quad (5)$$

Solutions to the respective time dependent forms of Equations 3, 4, and 5, appropriate for the particle target material geometry, can be found either by separation of variables, the use of Laplace or Fourier transformation techniques, or by standard numerical computational techniques or in combination.

We assume that the particles are uniformly distributed during production of the radioactive species. For the more general case, the distribution function S must be chosen to represent the actual distribution of the radioactive species within the target material. For a uniform distribution of particles such as assumed in the production of radioactive species with a primary ion beam of intensity I and charge Z , S is given by

$$S(x,t) = \sigma n \ell / ZeV \quad (6)$$

where e is the charge on the electron, n the number of interaction nuclei per unit volume, 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 of the irradiated sample.

For production of radioactive species with half-life $\tau^{1/2}$, E is given by

$$E(x,t) = n\lambda \text{ where } \lambda = 0.693/\tau_{1/2} \quad (7)$$

For solids, the diffusion process is dependent on the activation energy H_A required to move the atoms or vacancies from site to site is dependent upon H_A , and the temperature T according to:

$$D = D_0 \exp(-H_A/kT) \quad (8)$$

where D_0 is the intrinsic diffusion coefficient of the atom within the particular crystal matrix. D is related to the vibrational frequency and lattice parameters of the particular atom and crystal and k is Boltzmann's constant. H_A can be extricated from experimental data by measuring the dependence of D on target temperature T .

Figure 1 displays a computational simulation of the release of ^{58}Cu and ^{63}Cu from thin layers (5 μm) of Ni metal electroplated on Ir coated (1 μm) C fibers of diameter 6 μm as computed with DICTRA [6].

Fig. 1. Simulation of the diffusion release of ^{58}Cu from an annular deposit of Ni onto Ir coated, RVCF, similar to the composite target illustrated in Fig. 4 (Ni coating: 5 μm ; Ir coating: 1 μm ; C diameter: 5 μm). The target is assumed to operate at 1000°C . Diffusion is assumed to move radially outward due to a reflecting boundary condition imposed at the interface between the Ni deposit and the Ir coating.

Theory of Adsorption

Time delays, associated with adsorption processes, that are excessively long in relation to the life-time of the radioactive species can result in significant losses of beam intensity in an ISOL facility. The residence time of a particle on a surface is given by the Frenkel equation:

$$\tau = \tau_0 \exp[H_{ad}/kT] \quad (9)$$

where H_{ad} is the heat of adsorption or enthalpy required to evaporate the atom or molecule from the surface, k is Boltzmann's constant, T is the absolute temperature, and τ_0 is the time required for a single lattice vibration ($\sim 10^{-13} - 10^{-15}$ s). The heat of adsorption increases with increasing chemistry between the adsorbed atom and the surface where the adsorption takes place. This value varies widely depending upon the adsorbent/adsorbate combination.

The desorption rate of atoms per unit area dN/dt in thermal equilibrium with a surface at temperature T is given by

$$\frac{dN}{dt} = \frac{S(T)NkT}{h} \exp[(\Delta S - H_{ad})/kT] \quad (10)$$

where $S(T)$ is the temperature-dependent probability that the particle will stick to the surface (sticking coefficient), N is the number of atoms adsorbed per unit area, h is Planck's constant and ΔS is the change in entropy of the adsorbed particle.

Since it is desired to minimize the residence times of atoms/molecules on surfaces in the target/ion source,

the choice of the materials of construction for the vapor transport system of the source is extremely important. Coating the inner surfaces of the vapor transport tube (usually made of Ta or W) with a chemically inert material is expected to significantly reduce residence times and, therefore, the noble metals, iridium or rhenium, have been recommended for this use [7, 8].

NEW TARGETS AND TARGET CONCEPTS

New target concepts are presently under development at the HRIBF that, in principle, can be applied in the design and fabrication of targets with short diffusion lengths and high permeability properties required if present RIB facilities are to be successful in meeting the intensity and species needs of their respective research programs [9]. These new technologies include the choice of low-density target matrixes, such as carbon-bonded-carbon fiber (CBCF) or reticulated vitreous carbon fiber (RVCF), which can be used as the thermal transport structure for the removal of heat from the target material and as the plating matrix for the target material itself. Since short-lived particles must swiftly diffuse from the target material, the diffusion lengths must be short (thin target materials) and the target temperature must be as high as practicable. Techniques are presently available that can be used to uniformly deposit specified thicknesses of the material in question onto the support matrix of choice. At this point in time, several target coating schemes are being used or are under consideration for this purpose, they include: 1) chemical vapor deposition (CVD); 2) physical vapor deposition (PVD); electrolytic deposition (ED) and 3) sol-gel coating. In addition, cataphoretic or electrophoretic coating techniques are also under consideration as viable means for coating target matrix support structures with specified thicknesses of material.

Fibrous target materials.

In a few cases, fibrous materials with small fiber diameters are available. Figure 2 shows photographs of

Fig. 2. Photograph of an Al_2O_3 fibrous matrix (fiber diameter: 3 μm) which has been successfully used for the low-intensity, on-line production and release of $^{17,18}\text{F}$ as a means of evaluating the material for potential RIB generation.

Al_2O_3 fibrous materials. The Al_2O_3 fibrous target material has been successfully used to produce and release $^{17,18}\text{F}$ for potential use in the HRIBF research program [10]. As noted, the target matrix is highly permeable and, therefore, the release efficiency should be high.

Composite Target Matrix Designs

The objective of the target development program is to develop thin but highly permeable target matrix/heat-sink systems that, in combination, will permit operation of the target matrix at the maximum temperature allowable during production of radioactive species.

Reticulated-vitreous-carbon fibers (RVCF) and carbon-bonded-carbon (CBCF) fibers offer generic matrices for coating target materials. Figure 3 is a photograph of a carbon-bonded-carbon fiber (CBCF) fiber target matrix (fiber diameter: 6 μm) precoated with ~ 1

Fig. 3. Photograph of a carbon-bonded-carbon fiber (CBCF) matrix (fiber diameter: 6 μm) precoated with ~ 1 μm of Re followed by chemical vapor deposition (CVD) of 5 μm of SiC. The composite target matrix is highly permeable and will be tested for the release of short-lived P and S isotopes.

μm of Re followed by chemical vapor deposition (CVD) of 5 μm of SiC. The composite target matrix is highly permeable and will be tested for the release of short-lived P and S isotopes. Figure 4 displays electroplated Ni/RVCF composite target for future evaluation as a possible target material for use in generating ^{58}Cu . The

Fig. 4. Photograph of reticulated-vitreous-carbon-fiber (RVCF) matrix (fiber diameter: 54 μm) electroplated with ~ 3 μm Ni for potential use in generating ^{58}Cu .

choice of the particular target matrix system will depend on the availability of techniques for uniformly coating the target material onto the composite target matrix at the thicknesses required for efficient and fast diffusion release and the results of thermal transport studies [9].

The efficiency of the ionization process is strongly dependent on the pressure in the ionization chamber of the source and thus the vapor pressure of the target material; the vapor pressure of the target material is, in turn, strongly dependent on the target temperature; therefore, control and maintenance of a specific temperature, below a critical value, in the target holder/target matrix region of the target/heat sink assembly is critically important for successful and efficient generation of high intensity RIBs. We are presently designing a heat/sink system for mounting target matrices for control of target temperatures [9]. To achieve the primary objective of the project, the target/heat-sink assembly design will be optimized so that the particular target composite matrix can be maintained at a prescribed temperature which is limited by the maximum vapor pressure of the target material that can be tolerated before compromising the ion source efficiency and/or destroying the target through sublimation/vaporization processes. Research sponsored by the Oak Ridge National Laboratory, managed by Lockheed Martin Energy Research Corporation for the U.S. Department of Energy under contract Number DE-AC05-96OR22464.

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