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The diffusion properties of ion implanted species in selected target materials*

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ABSTRACT

Experiments important to the future success of the Holifield Radioactive Ion Beam Facility (HRIBF) are in progress at the Oak Ridge National Laboratory which are designed to select the most appropriate target material for generating a particular radioactive ion beam (RIB). The 25-MV HHIRF 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. The intensity versus time of implanted species, which diffuse from the high-temperature target material (~1700°C) and are ionized in the FEBIAD ion source, is used to determine release times for a particular projectile/target material combination. From such release data, diffusion coefficients can be derived by fitting the theoretical results obtained by computational solution of Fick's second equation to experimental data. The diffusion coefficient can be used subsequently to predict the release properties of the particular element from the same material in other target geometries and at other temperatures, provided that the activation energy is also known. Diffusion coefficients for Cl implanted into and diffused from CeS and Zr_5Si_3 and As, Br, and Se implanted into and diffused from Zr_5Ge_3 have been derived from the resulting intensity versus time profiles. Brief descriptions of the experimental apparatus and procedures utilized in the present experiments and plans for future related experiments are also presented.

1. INTRODUCTION

Many nuclear reactions important in the study of nuclear physics and astrophysics are experimentally inaccessible using stable beam/target combinations and, therefore, can only be studied with accelerated radioactive ion beams (RIBs). RIBs, therefore, offer unique opportunities to extend our knowledge of the nucleus and the energetics of our solar system. As a consequence, RIB facilities have been proposed or have been funded for construction in Asia, Europe, and North America [see, e.g., the facilities listed in Ref. 1]. Of the several facilities listed in Ref. 1, only the RIB facility at Louvain-la-Neuve is presently operational ,[2] the Holifield Radioactive Ion Beam Facility (HRIBF) at the Oak Ridge National Laboratory is now under construction,[3] and the RIB facility at GANIL has been recently funded.

Among several major challenges posed by generating and accelerating adequate intensities of RIBs, selection of the most appropriate target material is perhaps the most important because of the requisite fast and selective thermal release of minute amounts of the short-lived product atoms from the ISOL target in the presence of bulk amounts of target material. Satisfactory solutions to this challenging problem can be reached only by careful design of the ISOL

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source and by careful consideration of the physical, chemical, and metallurgical properties of the radioactive atom in relation to those of the target material. While there appears to be no lack of choices of sources for the efficient ionization of elemental or molecular feed materials, the primary problem lies in the availability and proper choice of target materials which are sufficiently refractory so that they can be heated to the elevated temperatures necessary for fast release of the product radioactive species without vaporization or sublimation of the target material itself. The vapor pressures of the unwanted species must be kept at a rate sufficiently low so as to avoid spoiling the rather stringent vacuum levels required for efficient ionization (~10^{-5} to ~10^{-4} Torr) [4,5]. Elements such as C, B, Cr, Hf, Ir, Mo, Nb, Os, Re, Ru, Ta, V, W, and Zr and several metal borides, carbides, nitrides, oxides, and a few sulfides which meet both the refractory temperature and low vapor pressure requirements, when appropriate, can be used for target materials. In addition, several intermetallic alloys such as Hf_3Si_2, $LaAl_2$, Zr_5Ge_3, and Zr_5Si_3, as well as metal aluminates, silicates, titanates, and other more complex compounds also meet these criteria and, therefore, can be considered as potential target materials. Obviously, compounds which contain the highest percentage of the element of interest are desirable so as to maximize the production rate of the radioactive species of interest. Still other factors which limit the choice of target materials must be considered such as the production of unwanted, long-lived radioactive by-products. Ideally, the target material should be selected so as to maximize the speed of release of the species of interest at the maximum operational temperature which avoids spoiling the efficiency of the source. More than 68 of the elements in the periodic chart have been produced, released, mass separated, and their products studied at various ISOL facilities. For example, at the CERN-ISOLDE facility, more than 600 radioactive beams representing 66 elements have been provided for research with intensities ranging up to 10^{12} ions/s.[6]

Fast release of a short-lived radioactive species from a target material most generally requires high temperature target/ion source operation. In the ideal case, the target material should be refractory while simultaneously exhibiting low vapor pressures (<10^{-4} Torr) at the elevated operational temperatures of the target-ion source (~1300°C to ~2000°C) in order to avoid sublimation or vaporization of the target to pressures which exceed acceptable values for efficient ionization. The choice of target material is further restricted by the requirement that the radioactive material itself not be refractory and that it not form refractory compounds within the target material. The radioactive atoms should, in the ideal case, possess physical and chemical properties almost opposite to those of the target material; that is, they should be easily diffused from the target material either in elemental form or in compound form, and upon exit, be readily desorbed and vaporized from the target surface. These idealized differences in chemical and physical properties which are desirable for fast release of the species of interest are not often practically realizable, particularly for close-lying elements where their physical and chemical properties are often similar. This poses particular challenges for present and future RIB facilities.

Ion implantation, in principle, offers a cost effective and powerful technique for predetermining the time-release behavior of stable complements of interesting radioactive species from target materials which are candidates for their generation, as well as a straightforward method for determining diffusion coefficients for many element/target combinations. Experiments are under way at the HRIBF[7] which utilize HHIRF tandem accelerator beams to implant stable complements of interesting radioactive atoms into refractory targets mounted version of the FEBIAD ion source[8] of the "on-line" isotope separator (UNISOR).[9] The ion implantation techniques utilized to select the target material are similar to those described by Kirchner.[10] In this report, we describe experimental equipment, procedures, and results derived from implanting Cl into CeS and Zr_5Si_3, and As, Br, and Se into Zr_5Ge_3. Release times and diffusion coefficients will be presented for each of these species. The diffusion coefficients will also be used to model the release of radioactive ion beams from homogeneously distributed planar- or macro-spherical-geometry targets which represent the actual distributions and target geometries which will be used for RIB generation at the HRIBF;[3] examples of the predicted time-release profiles from such targets are also presented.

2. PROCESSES WHICH AFFECT RELEASE TIME

The principal means whereby radioactive species are lost between initial formation and utilization are associated with diffusion and surface adsorption processes, as well as efficiencies for forming beams of the species. The diffusion and surface adsorption processes are briefly described below.

2.1 Diffusion Theory

A binary diffusion mechanism must be responsible for the time-temperature dependent release of a nuclear reaction product species embedded in a chemically dissimilar target material (see, e.g., Ref. 11 for a discussion on diffusion processes). The atoms and vacancies created in the nuclear reaction process rearrange themselves within the solid through diffusion until equilibrium is reached in the presence of a concentration gradient of impurity atoms or vacancies. 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:

$$J = -D\nabla n \tag{1}$$

where D is the diffusion coefficient. The time-dependent form of Eq. 1 is known as Fick's second equation. The one-dimensional form is expressed as follows:

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

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

$$\frac{\partial n}{\partial t} = \frac{D}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial n}{\partial r}\right) - \lambda n \tag{3}$$

where $\lambda = 0.693/\tau_{1/2}$.

The λn term is included to account for losses with time when the atom is radioactive with half life $\tau_{1/2}$. The distribution function of the ion implanted species is sharply Gaussian and localized in a very discrete position beneath the target surface. The depth of implantation depends on the projectile, the projectile energy, and atomic number of the constituents which make up the target material, and the density of the target material. Fig. 1 displays a TRIM[14] simulation of the final distribution of a 209-MeV ^{80}Se ion beam in Zr_5Ge_3 after passing through a Ta window and slowing down and stopping in the target material; the Ta window degrades the initial beam energy from 270 MeV. As noted, the distribution of particles is sharply defined. The initial distribution of the implanted species affects the time release profile of the implanted species.

The diffusion process is driven not only by the concentration gradient, but by the activation energy H_A required to move the atoms or vacancies from site to site. The diffusion coefficient is dependent upon H_A, carrying the temperature dependence given by:

$$D = D_o \exp(-H_A/kT) \tag{4}$$

where D_o 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. Values for D_o can be estimated from experimental data provided that H_A is known. H_A can be extricated from experimental data by measuring the dependence of D on target temperature T; the present version of the FEBIAD ion source precludes such measurements, since the target temperature cannot be varied. However, H_A can be assigned reasonable values which enables the prediction of the release function for other temperatures. We have arbitrarily assigned H_A a value of 1.0 eV for estimating the release of Cl from CeS operated at other temperatures, strictly for purposes of illustrating the capability of the modeling process. This diffusion coefficient D, extricated from experimental information, can be utilized for predicting the release times for other distributions of the species from the same target material at the same temperature. As noted from Eq. 4, higher temperatures significantly affect release times of particles from the solid.

2.2 Theory of Adsorption

Any time delays 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. Upon diffusion, the atom of interest must be evaporated from the target source and effused into the ionization chamber of the ISOL source. The residence time of a particle on a surface is given by the Frenkel equation:

$$\tau = \tau_o \exp[H_{ad}/kT] \tag{5}$$

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_o is the time required for a single lattice vibration ($\sim 10^{-13}$ 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. In our model, the heat of adsorption was assumed to have a constant value for all species of 3.0 eV. This value is relatively well defended in the literature. [12]

The desorption rate per unit area, $\partial N/\partial t$ of adsorbed atoms in thermal equilibrium with a surface at temperature T can be given by

$$\frac{\partial N}{\partial t} = S(T)N\frac{\omega_o}{2\pi}\exp[-H_{ad}/kT] \tag{6}$$

where S(T) is the probability that the particle will stick to the surface (sticking coefficient) which is a temperature-dependent, N is the number of atoms adsorbed per unit area, and $\omega_o/2\pi = 1/\tau_o$.

Since it is desired to minimize the residence times of atoms/molecules on surfaces in the target/ion source, the choice of materials for high-temperature vapor transport source components is extremely important. Coating the inner surfaces of the ion source (usually Ta or W) with a chemically inert material with which the vapor comes in contact during transport from the target to the ionization chamber is expected to significantly reduce residence times of particles on the surface and, therefore, iridium or iridium-coated materials such as tantalum or tungsten have been recommended for use.[7] Iridium, though expensive, is especially desirable; for example, the heat of adsorption for oxygen on tantalum is ~9.74 eV,[13] which corresponds to a residence time of 3.7×10^{8} seconds at 2273°K, whereas the heat of adsorption for oxygen on iridium is ~3.33 eV,[13] which correlates to a resident time of 2.4×10^{-6} seconds at the same temperature. The ratio τ_{Ta}/τ_{Ir} is 1.5×10^{14}. The residence times for chemically active materials will be essentially negated provided that a stable, long-lived surface coating can be effected on all internal source components with which the vapors come in contact.

3. EXPERIMENTAL APPARATUS AND PROCEDURES

The 25-MV tandem accelerator is utilized to implant stable complements of interesting radioactive atoms into refractory targets maintained in the on-line FEBIAD ion source which is the standard source used by the UNISOR facility; A schematical representation of the UNISOR facility utilized in all of the diffusion release measurements is displayed in Fig. 2. In the ISOL scheme, the primary ion beam intercepts the target which resides either in the source itself or is closely coupled to the ion source by means of a vapor transport tube. The FEBIAD ion source is schematically displayed in Fig. 3.

The experiments described in this paper were performed by implanting Cl^{8+} into CeS, Zr_5Si_3, and As^{13+}, Br^{13+}, and Se^{13+} ions implanted into Zr_5Ge_3 targets maintained in the anode structure of the FEBIAD ion source displayed in Fig. 3. The FEBIAD ion source differs from conventional plasma-discharge sources in that its operational pressure range lies lower than the minimum pressure required to achieve a stable plasma discharge. The source is suitable for ISOL applications involving the use of heavy ions to produce the radioactive species of interest in that it operates stably and efficiently over a pressure range of $\sim 1 \times 10^{-5}$ to $\sim 2 \times 10^{-4}$ Torr at elevated temperatures.[4] For a more thorough description of the FEBIAD ion source in relation to other high-efficiency sources, see e.g., Ref. 15. Before entering the target, which is maintained at anode potential in the FEBIAD ion source, the ion beam passes through a thin window of some heavy elemental material, such as tantalum, which slightly degrades the energy of the beam. The thickness of the window is typically 3.5 to 5 mg/cm^2. The final energy after passing through the window is chosen so that the ions penetrate to a depth of 2 to 35 μm in a thin circular disk (~9 mm in diameter and 1.5 mm in thickness) of the material of interest. The simulation program, TRIM,[14] which calculates the energy loss of the projectile while slowing down in the target material, is used to numerically determine the position and distribution of the implanted species within the target material. A simulation program, called DIFFUSE,[16] is used to solve Fick's second equation (Eq. 2) and to determine release curves for Cl diffusing from CeS and Zr_5Si_3, and As, Br, and Se diffusing from Zr_5Ge_3. The input parameters, required by DIFFUSE to fit to the experimental data, include the particle distribution function as calculated by the use of TRIM, the activation energy, and the temperature T of the sample. Initial estimates of D are determined by fits to the data in the tail of the "beam-off" part of the intensity versus time release curve. D is then iteratively altered until an acceptable fit is found.

Upon deposition within the target, the neutral species diffuse randomly in the high-temperature sample, which is maintained at a temperature of ~1645°C prior to implantation. Beam heating effects raise the target temperature in proportion to the beam power; this effect has been measured; typically a 10-Watt beam raises the temperature by an additional 50°C. The target thus resides at ~1700°C during the implantation. Particles which diffuse out of the face of the beam entry side of the target are evaporated from the target surface and are transported into the anode region of the ion source through effusive flow. After passing into the anode structure, they are bombarded with an electron beam accelerated from a hot cathode. A fraction of the particles are ionized, extracted from the ion source, mass analyzed, and the signal versus time of the mass selected species is recorded with the "beam-on-target"; the signal rises according to the release rate of the implanted species and reaches saturation or a constant value, at which time the beam is removed from the target. During the "beam-off-target" time period, the intensity decreases with time and is monitored until the signal reaches background levels. The time required for releasing 50% of the saturation level is defined as the release time for the particular species from the target material.

4. RESULTS

4.1 Experimental Time-Release Profiles for Ion-Implanted Species in Planar-Geometry Targets

The time-release information derived from ion implanted targets, which have sharply Gaussian distributions, is only of qualitative value for estimating release of radioactive species from RIB target materials where the nuclear reaction products are more or less uniformly distributed within the sample. The release times from implanted targets are dependent on the initial distribution, as well as the depth of the implanted material. Therefore, information derived from such experiments cannot be directly used to predict release times of the complementary, short-lived, homogeneously distributed, radioactive species. However, diffusion coefficients D can be derived by fitting solutions to either Eq. 2 or Eq. 3, whichever is most appropriate, to experimental release data; the diffusion coefficients can then be used to predict release times of short-lived radioactive complements of the implanted species from homogeneously distributed, planar- or spherical-geometry samples at the same temperature which are the principal target geometries that will be used for actual RIB generation. The experimentally measured release profiles reflect the total accumulated time required for diffusion τ_d, effusion τ_e, and ion transport τ_s to the detector system following mass analysis. The diffusion coefficients and release times derived from the data are only valid whenever $\tau_d >> \tau_e + \tau_s$ or whenever τ_e and τ_s are known. While τ_s is always precisely known, τ_e must be estimated or measured independently.

Fig. 4 compares the experimentally measured time-release profiles of ^{37}Cl implanted to a depth of 2.6 μm in CeS (target temperature: 1690°C) with fits to the data derived by use of DIFFUSE. The dashed line superposed on the experimental data represents a computational fit to the data using the program DIFFUSE from which the diffusion coefficient D was determined. As noted, the fits to the release profile are exceptionally good. Figure 5 displays theoretical "beam-on-target" and "beam-off-target" time-release spectra for ^{37}Cl and ^{33}Cl diffusing from homogeneously distributed, 5-μm-diameter spherical-geometry macro-particles of CeS at 1690°C and 1990°C as predicted by use of the computer code DIFFUSE (Ref. 16). The code was utilized to determine the diffusion coefficient D by iteratively fitting solutions to Fick's second equation (Eq. 2) to the experimental data taken at 1690°C displayed in Fig. 4. The predictions made for the diffusion release at a target temperature of 1990°C were estimated by assuming an activation energy of 1.0 eV which correlates with an intrinsic diffusion coefficient, $D_o = 1.6 \times 10^{-7}$

cm^2/s. The diffusion coefficient D at the temperature for which the release measurements were made is D = 4.4 x 10^{-10} cm^2/s. We have assumed an activation energy of H_A in Eq. 3 of H_A = 1.0 eV for purposes of illustrating the behavior of the release of Cl from CeS at higher temperatures than those which were used in the ion implantation experiments. This value may be too low, but is reasonable in relation to measured values commonly found in the literature (see, e.g., Ref. 11).

The time-release behaviors of As, Se, and Br implanted into Zr_5Ge_3 targets and Cl implanted into Zr_5Si_3 targets were measured using the apparatus and procedures discussed in Section 3. These target materials are very refractory, having melting points from 2100°C to 2300°C and are candidates for use in generating the radioactive complements of these species. These data were also analyzed and diffusion coefficients extricated. In general, several minutes of ion bombardment are found necessary to "clean up" the target. Figures 6-9 display, respectively, the experimentally measured time-release profiles for Cl implanted into and diffused from Zr_5Si_3, and As, Br, and Se implanted into and diffused from Zr_5Ge_3 samples; the solid lines represent fits to the experimental data. As noted, the fits provide good to excellent agreement with the corresponding experimental data. From fits to these data, diffusion coefficients D were derived which were then used to predict the time release profiles from homogeneously distributed macro-sphérical or planar targets (Figs. 9-13) at the same temperature, geometries which will be used most often for RIB generation. The release time τ is defined as the time required to release 50% of the steady-state ion beam intensity diffused from the target.[9] The diffusion coefficients, ranges of implantation, and release times for these projectile/target combinations are listed in Table I.

4.2 Predicted Time-Release Profiles for Homogeneously Distributed Species in Planar and Spherical Targets

During operation of the HRIBF, the radioactive species will be uniformly distributed within either planar or spherical targets. The release times for uniformly distributed species will differ from those obtained from the Gaussian distributions characteristic of ion implanted samples and, therefore, the release time information derived from ion implanted samples is only of qualitative value for use in predicting release times from actual RIB target materials. However, diffusion coefficients can be extricated from such data and used to predict release times for other target geometries and at different target temperatures. In order to use solutions to Fick's equation (Eqs. 2 or 3) for making predictions at temperatures other than those used in the ion implantation experiments, the activation energy must be known.

Examples of the predicted releases of stable and complementary radioactive species from homogeneously distributed spherical or planar target materials are shown in Figs. 10-13. The target material for Cl is assumed to be Zr_5Si_3, while that for Br, As, and Se is assumed to be Zr_5Ge_3. The release profiles for the 20-µm-thickness planar-disk targets displayed in Figs. 10-13, which are, in fact, too thin to fabricate in practice, serve only as a means of comparison of the planar disk targets with the practically sized, 20-um spherical-geometry targets. It is obvious that spherical macro-particle target materials are always desirable if one can avoid sintering the materials during the release process. The actual release times for radioactive particles which have been released by diffusion from spherical particles will be moderated by the hold-up times associated with the times required to migrate through the distribution of material. These times may be long enough to offset any advantage gained by using spherical particles coupled with the fact that the target must be operated at temperatures below the sintering temperature (~80% of the melting point of the particular material). Table II summarizes the predicted release times for each of the species homogeneously distributed spherical- or planar-geometry targets.

5. 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 ion beams. Release time and efficiency information derived from such experiments can be used to realistically estimate beam intensities which could be produced in a particular ISOL ion source during RIB beam generation. Diffusion coefficients extracted from this 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. The method also offers a fast, simple, and cost-effective means for determining diffusion coefficients for element/solid target material combinations which are often time consuming, expensive, and tedious to experimentally derive by other means. Future ion implantation, diffusion-release experiments will be conducted by use of a CERN-type RIB target/ion source which will enable variation of the target temperature. This capability will enable determination of activation energies H_A.

6. ACKNOWLEDGMENTS

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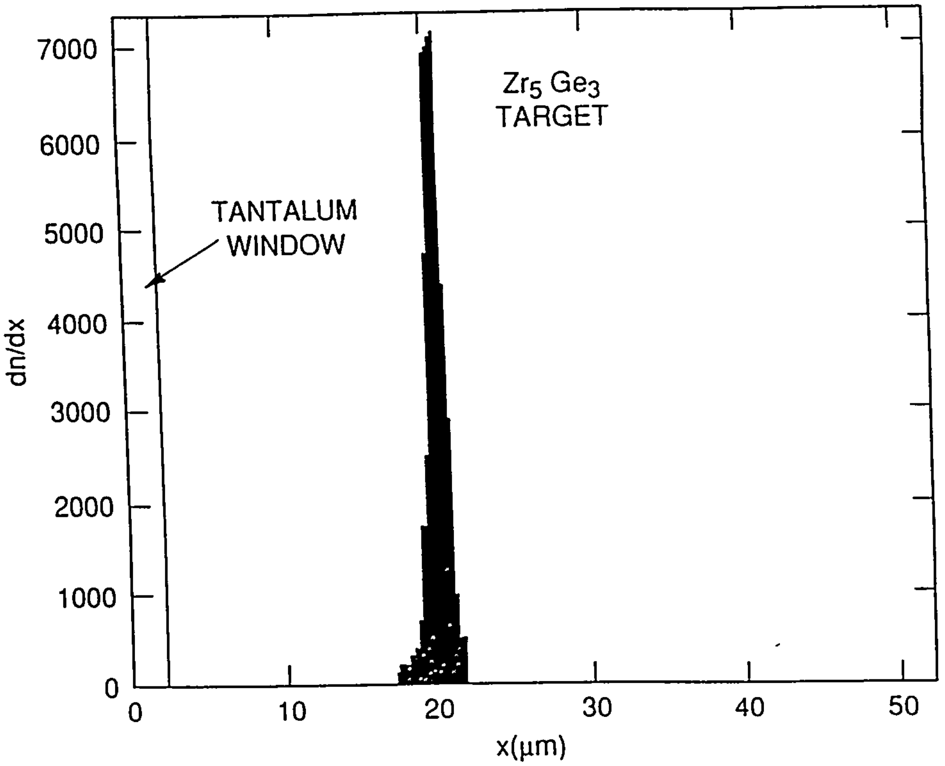

Fig. 1. TRIM simulation of a 270-MeV ^{80}Se ion beam implanted into Zr_5GE_3 through a 2-μm Ta window.

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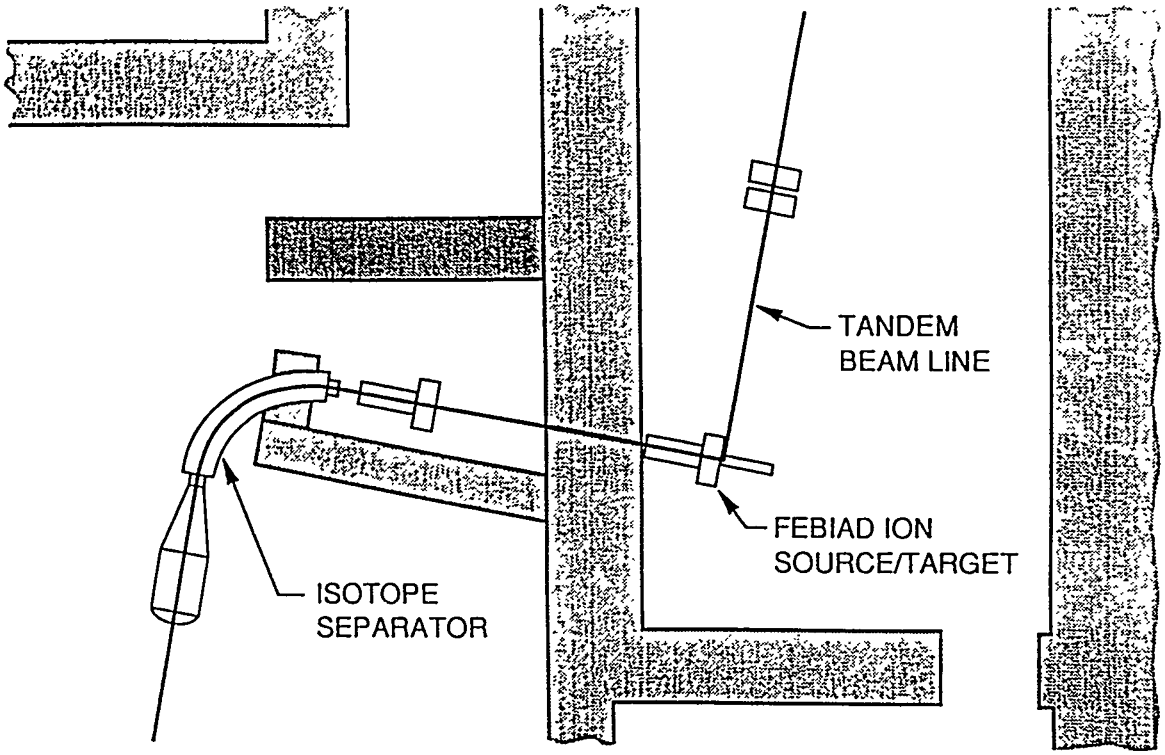

Fig. 2. Schematic representation of the UNISOR facility utilized in the present element/refractory target release experiments.

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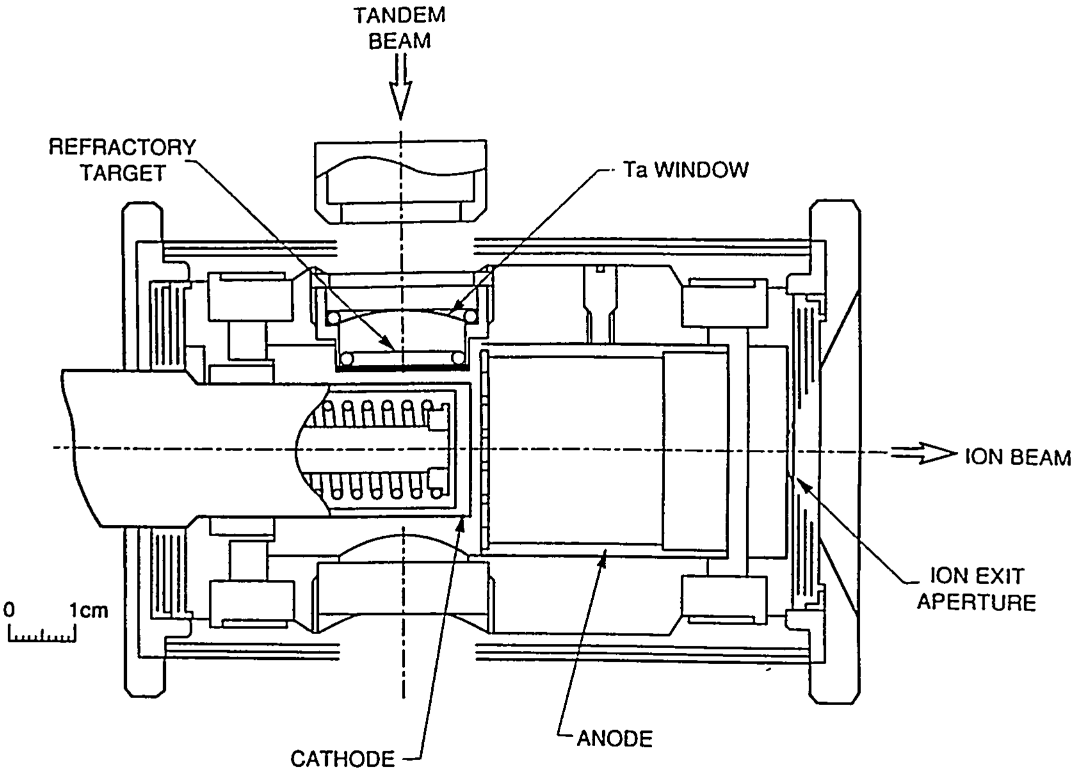

Fig. 3. Schematic drawing of the FEBIAD ion source used in the element/refractory target release experiments.

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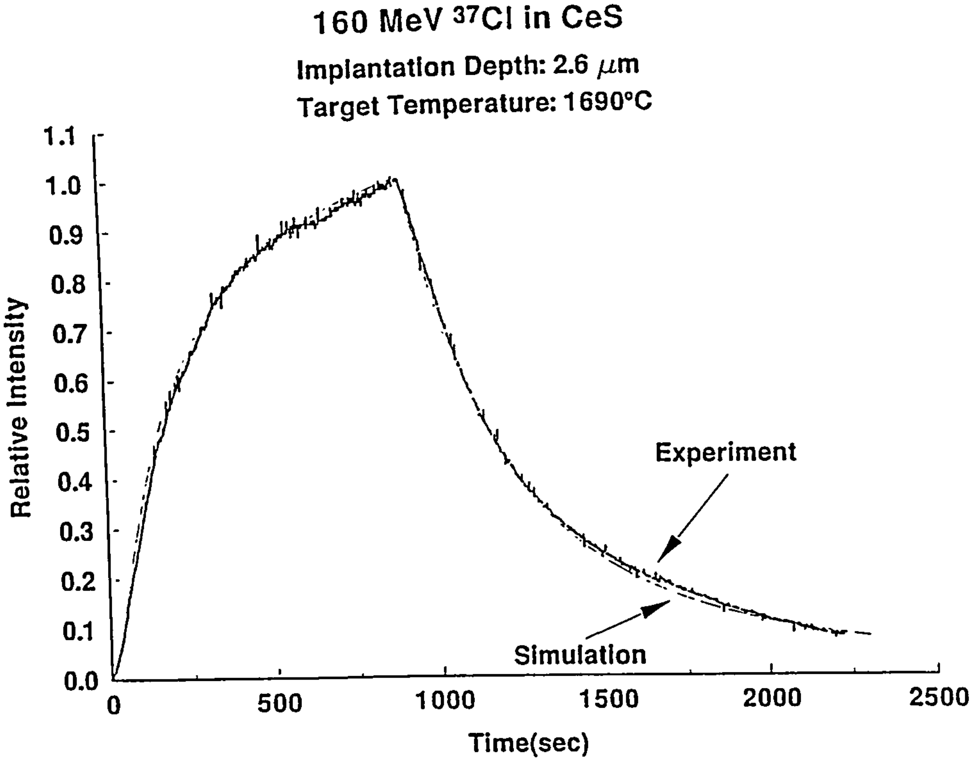

Fig. 4. Typical "beam-on-target" and "beam-off-target" time release spectra for ^{37}Cl implanted into and released from planar geometry CeS targets. The dashed line represent a fit to the experimental data, derived by iteratively solving Fick's second equation (Eq. 2) by the use of the computer code, DIFFUSE (Ref. 16). Target temperature: 1690°C; Implantation depth: 2.6 μm; Diffusion coefficient: $D = 4.4 \times 10^{-10}$ cm^2/s.

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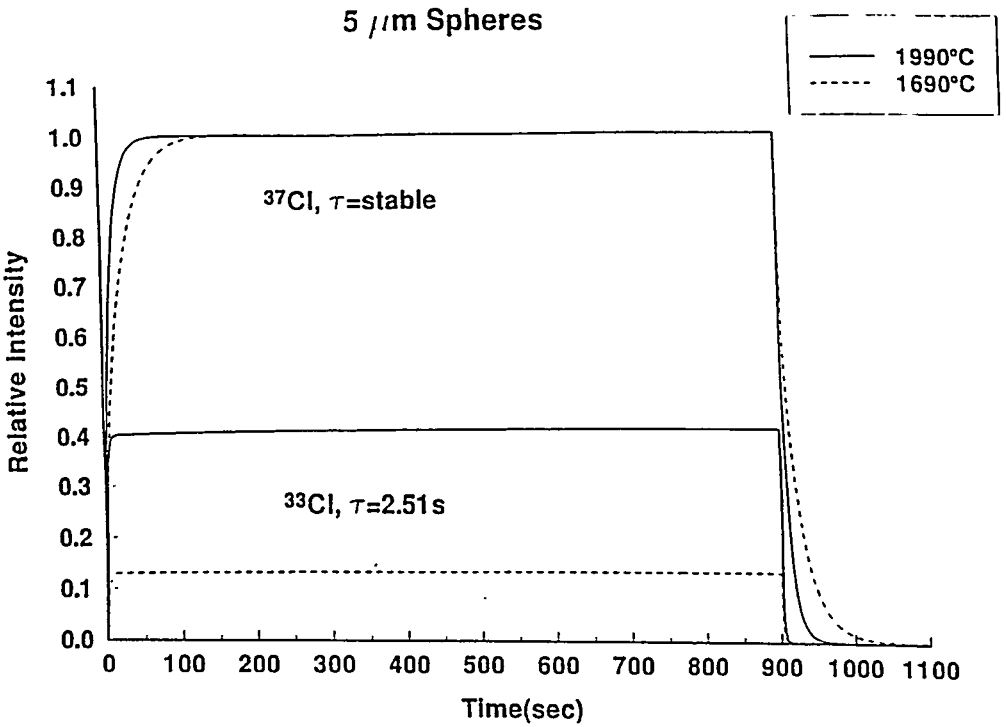

Fig. 5. Theoretical "beam-on-target" and "beam-off-target", time-release spectra for ^{37}Cl and ^{33}Cl diffusing from homogeneously distributed, 5-μm-diameter spherical-geometry macro-particles of CeS at 1690°C and 1990°C as predicted by use of the computer code DIFFUSE (Ref. 16). The code was utilized to determine the diffusion coefficient D by iteratively fitting solutions to Fick's second equation (Eq. 2) to the experimental data taken at 1690°C displayed in Fig. 4. The predictions, made for the diffusion release at a target temperature of 1990°C, were estimated by assuming an activation energy of 1.0 eV which correlates with an intrinsic diffusion coefficient, D_0 = 1.6 x 10^{-7} cm^2/s. Diffusion coefficient at a target temperature of T = 1690°C: D = 4.4 x 10^{-10} cm^2/s.

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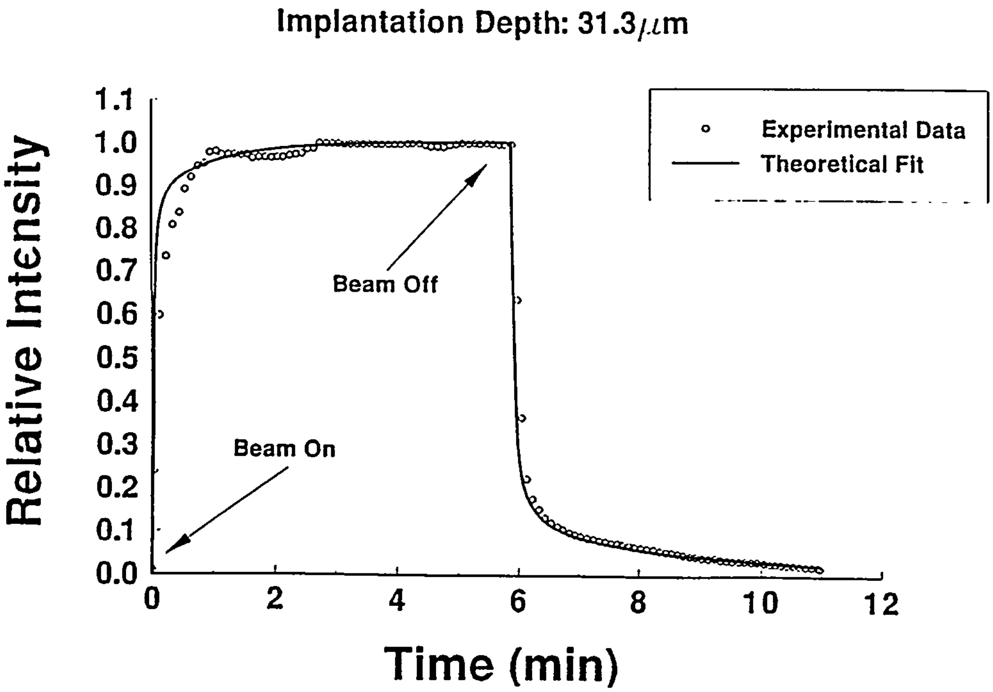

Fig. 6. Typical "beam-on-target" and "beam-off-target" time-release spectra for ^{35}Cl implanted into and diffused from Zr_5Si_3 planar geometry targets. The solid line represents a fit to the experimental data derived by iteratively solving Fick's second equation (Eq. 2) by use of the computer program DIFFUSE (Ref. 16). Target temperature: 1695°C; Diffusion coefficient: D = 1.2 x 10^{-5} cm^2/s.

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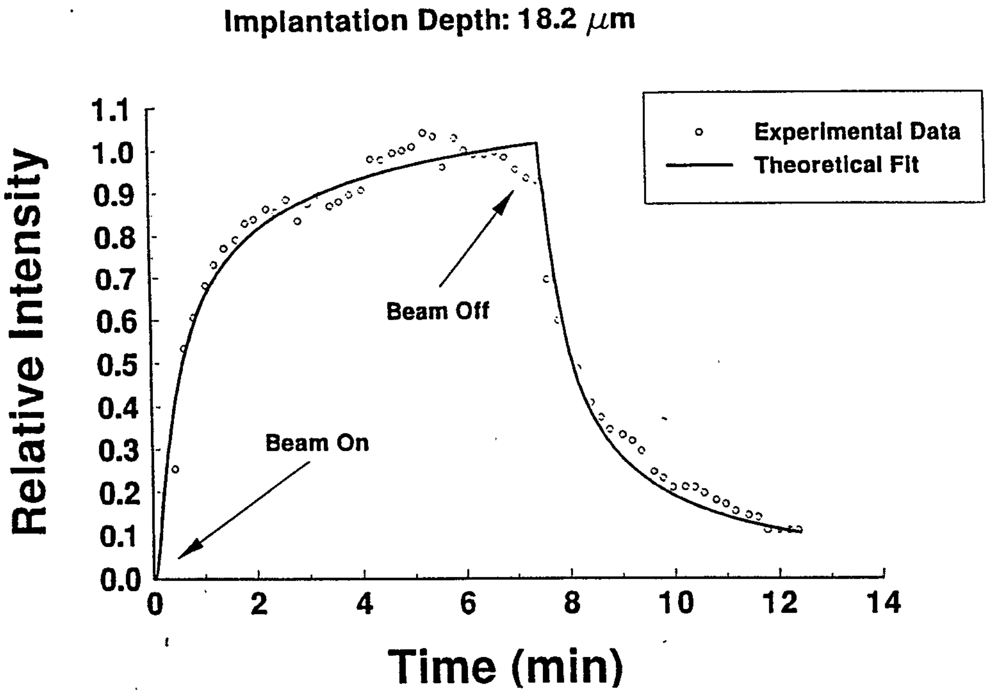
Implantation Depth: 18.2 μm

Fig. 7. Typical "beam-on-target" and "beam-off-target" time-release spectra for ^{75}As implanted into and diffused from Zr_5Ge_3 planar geometry targets. The solid line represents a fit to the experimental data derived by iteratively solving Fick's second equation (Eq. 2) with the computer program, DIFFUSE (Ref. 16). Target temperature: 1670°C; Diffusion coefficient: $D = 5.1 \times 10^{-8}$ cm^2/s.

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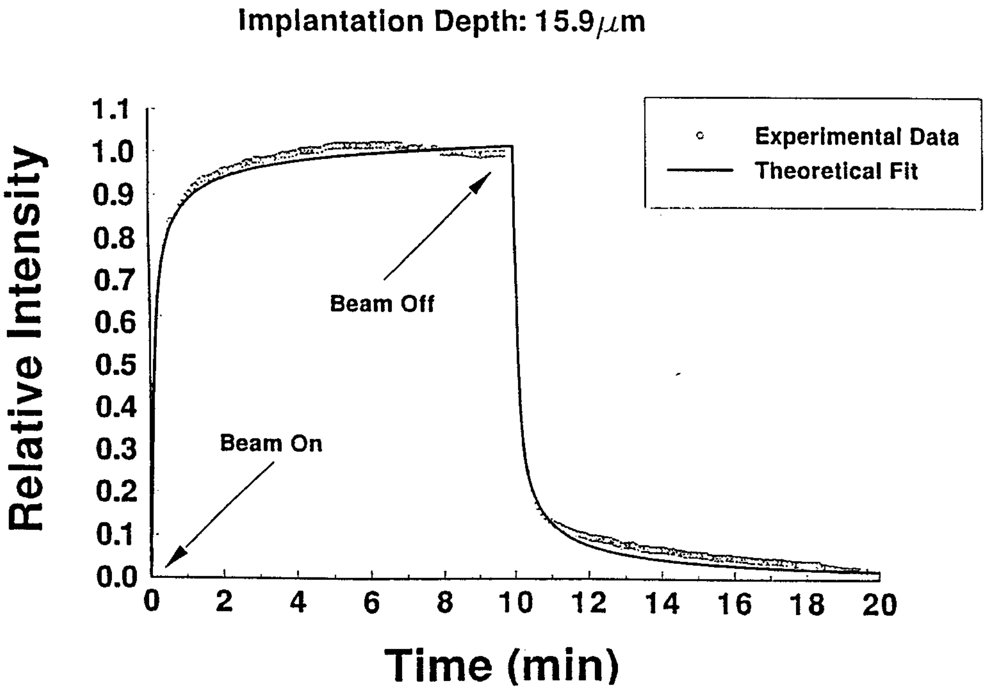
Implantation Depth: 15.9μm

Fig. 8. Typical "beam-on-target" and "beam-off-target", time-release spectra for ^{79}Br implanted into and diffused from planar geometry Zr_5Ge_3 targets. The solid line represents a fit to the experimental data, derived by iteratively Fick's second equation (Eq. 2) by use of the computer program, DIFFUSE (Ref. 16). Diffusion coefficient: $D = 5.2 \times 10^{-7}$ cm^2/s; Target temperature: 1675°C.

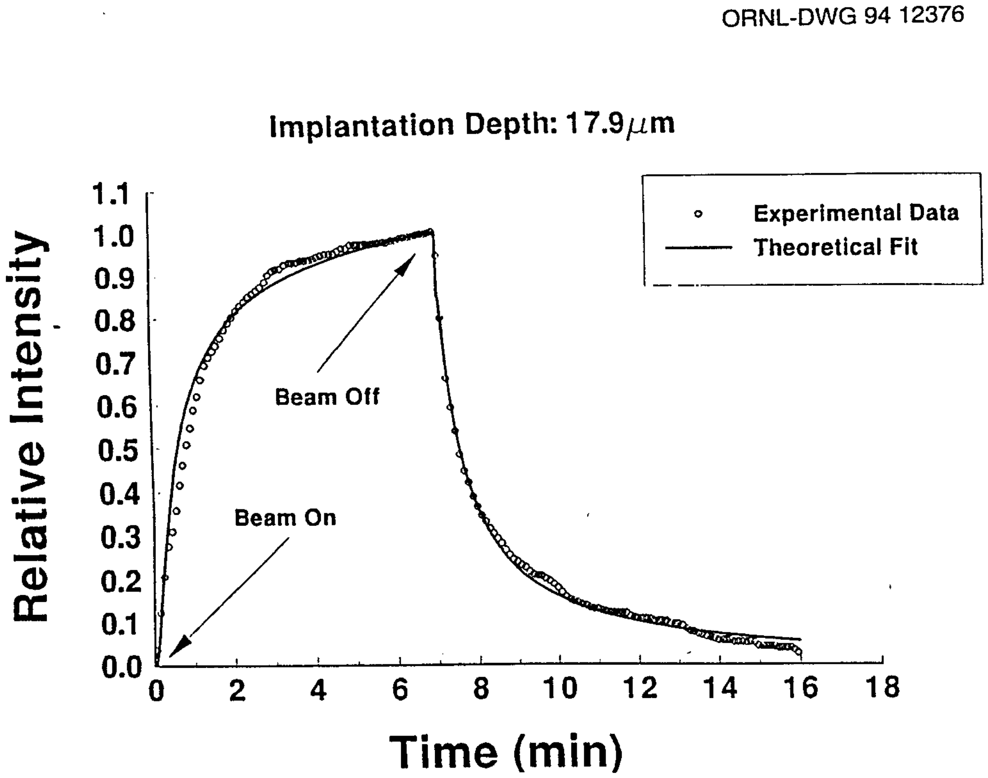

Fig. 9. Typical "beam-on-target" and "beam-off-target, time-release spectra for ^{78}Se implanted into and diffusing from planar geometry Zr_5Ge_3 targets. The solid line represents a fit to the experimental data, derived by iteratively solving Fick's second equation (Eq. 2) by use of the computer program DIFFUSE (Ref. 16). Target temperature: 1670°C; Diffusion coefficient: $D = 5.1 \times 10^{-8}$ cm^2/s.

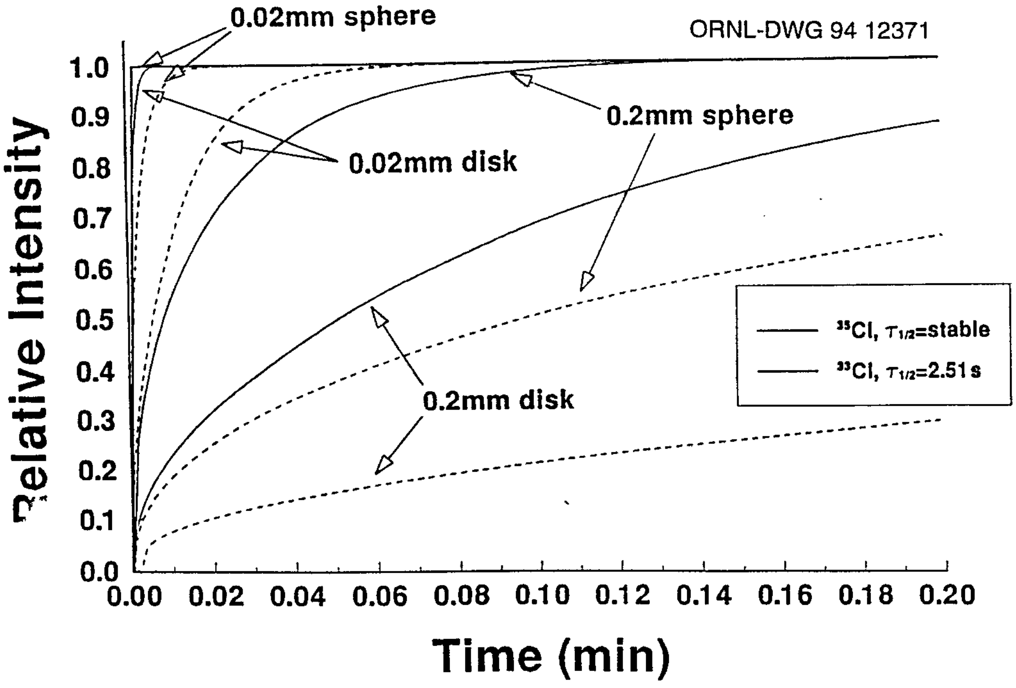

Fig. 10. Theoretical "beam-on-target", time-release spectra for homogeneously distributed ^{35}Cl and ^{33}Cl diffusing from 20-μm and 200-μm-diameter spherical-geometry and 20-μm and 200-μm-thick planar-geometry Zr_5Si_3 targets as predicted by use of the computer program DIFFUSE (Ref. 16). Target temperature: 1695°C; Diffusion coefficient: $D = 1.2 \times 10^{-5}$ cm^2/s.

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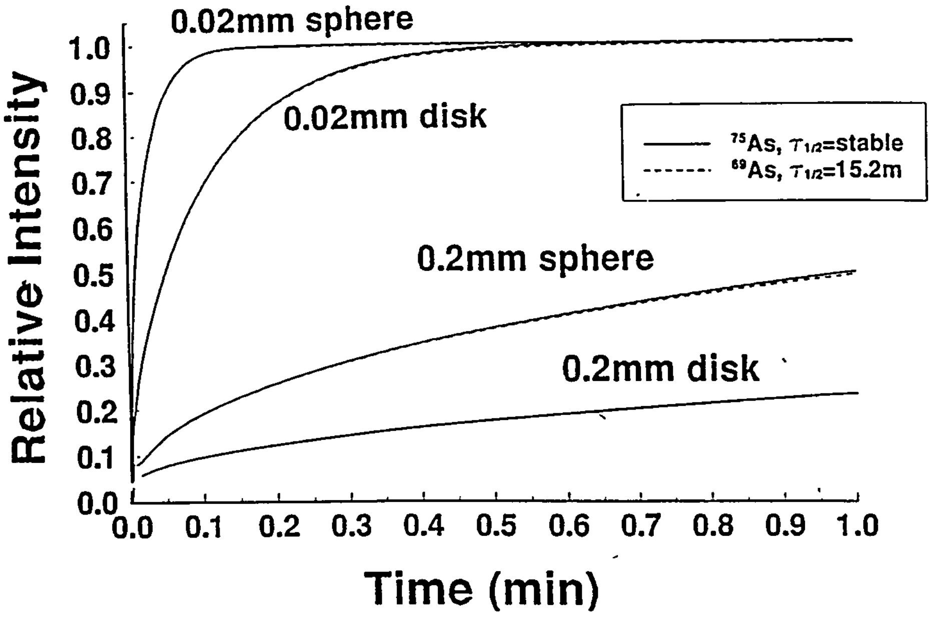

Fig. 11. Theoretical "beam-on-target", time release spectra for homogeneously distributed ^{79}As and ^{69}As diffusing from 20-µm and 200-µm-diameter spherical-geometry and 20-µm and 200-µm-thick planar-geometry Zr_5Ge_3 targets as predicted by the use of the computer program DIFFUSE (Ref. 16). Target temperature: 1670°C: Diffusion coefficient: $D = 5.1 \times 10^{-8}$ cm^2/s.

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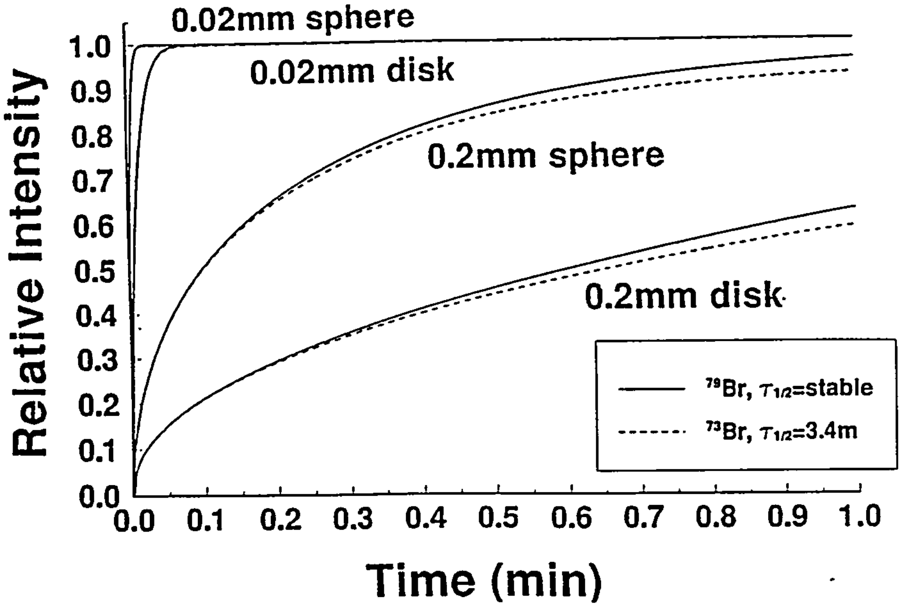

Fig. 12. Theoretical "beam-on-target", time-release spectra for homogeneously distributed ^{79}Br and ^{73}Br diffusing from 20-µm and 200-µm spherical-geometry and from 20-µm and 200-µm-thick planar-geometry Zr_5Ge_3 targets as predicted by the use of the computer code, DIFFUSE (Ref. 16). Target temperature: 1675°C; Diffusion coefficient: $D = 5.2 \times 10^{-7}$ cm^2/s.

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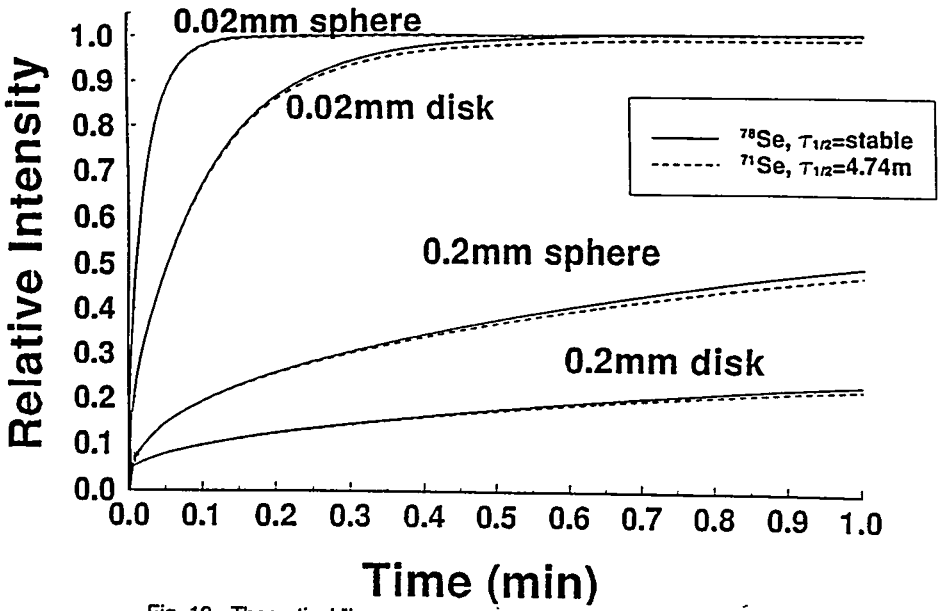

Fig. 13. Theoretical "beam-on-target", time-release spectra for homogeneously distributed ^{78}Se and ^{71}Se diffusing from 20-µm and 200-µm spherical geometry and 20-µm and 200-µm-thick planar-geometry Zr$_5$Ge$_3$ targets as predicted by the use of the computer program DIFFUSE (Ref. 16). Target temperature: 1670°C; Diffusion coefficient: $D = 5.1 \times 10^{-8}$ cm^2/s.