Simulation of the Effusive-Flow of Reactive Gases in Tubular Transport Systems: Radioactive Ion Beam Applications

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Maximum practically achievable intensities are required for research with accelerated radioactive ion beams (RIBs). Time delays due to diffusion of radioactive species from solid or liquid target materials and their effusive-flow transport to the ion source can severely limit intensities of short-lived radioactive beams, and therefore, such delays must be minimized. An analytical formula has been developed that can be used to calculate characteristic effusive-flow times through tubular transport systems, independent of species, tube material, and operational temperature for ideal cases. Thus, the equation permits choice of materials of construction on a relative basis that minimize transport times of atoms or molecules moving through the system, independent of transport system geometry and size. In this report, we describe the formula and compare results derived by its use with those determined by use of Monte-Carlo techniques.

I. INTRODUCTION

The Isotope Separator On-Line (ISOL) technique is most frequently used to produce short-lived isotopes for research at radioactive ion beam (RIB) facilities such as the HRIBF [1]. After being created in the matrix of a solid or liquid target, short-lived species must diffuse from the target material and then be transported, in gaseous or vapor form, through the transport system to an ion source where they are ionized and accelerated. If the integrated time for these processes is long with respect to the lifetime of the species, the intensity of the RIB will be seriously compromised. Thus, it is desirable to minimize the times associated with both processes. Consequently, it is important to know in advance, the delay times of these radioactive species in the combined target-material/vapor transport system. Given the diffusion coefficient for the species/target material binary combination, the time dependence of the diffusion process can be determined by solving Fick's second equation [2]. The Monte-Carlo statistical technique [3] can be used to determine characteristic effusive-flow transport times of atoms or molecules through simple transport systems. Here-to-fore, no equivalent analytical method has been developed that can be used in lieu of the Monte-Carlo statistical technique. In this report we describe an equation that can be used to extricate characteristic effusive-flow times for chemically active atoms/molecules through simple tubular vapor transport systems.

II. COMPUTER SIMULATIONS

The Monte-Carlo code, *Effuse* [3], is used to validate the accuracy of our analytical model. *Effuse* follows individual particles, from a statistically significant ensemble of particles (e.g., 10000), through the vapor-transport-system by randomly choosing starting parameters at entrance to the vapor-transport tube (e.g., starting angles and radial positions) and at positions along the tube for particles leaving surface sites during desorption, after residing on the walls of the tube for characteristic residence times, τ_{ad}. The characteristic time for transport is derived from a plot of the time distributions for all particles moving through the system. For each simulation, the code calculates the average distance traveled per particle, L, and the average number of interactions with the walls of the tube (bounces), N_b, for a set of given tube parameters (radius: a; length: l).

III. THE EFFUSIVE-FLOW FORMULA

For an ideal gas in a tube of radius, a, at low pressure, the steady-state flow-rate, dN/dt, for particles with average velocity, v, flowing through a tube under a density gradient, dn/dz, is given by the familiar relation,

• Managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract DE-AC05-00OR22725.

$$\frac{dN}{dt} = -\left\{\frac{2\pi a^3}{3}\right\}\upsilon\frac{dn}{dz} = -\left\{\frac{2\pi a^3}{3k_B T}\right\}\upsilon\frac{dp}{dz} \tag{1}$$

taken from the kinetic theory of gases [4]. In Eq. 1, n is the particle density; υ is the velocity of the atoms or molecules; and k_B is Boltzmann's constant. By solving the time dependent form of this equation, the following time distribution of particles in the tube at time, t, for chemically active radioactive particles with lifetime, $\tau_{1/2}$, can be derived:

$$N = N_0.Exp(-\lambda t).\exp\Big(-t\Big/\Big(3/4\big\{N_B\tau_0.\exp(-H_{ad}/k_B T) + L/\upsilon\big\}\Big)\Big) \tag{2}$$

where N_0 is the number of particles in the volume at time, t = 0, $\lambda = 0.693/\tau_{1/2}$; H_{ad} is enthalpy of adsorption with $\tau_0 \cong 3.4.10^{-15}s$. The factor 3/4 in the denominator of Eq. 2 was obtained by comparing results derived from the equation with those derived by use of *Effuse*. As noted in Eq. 2, both *L (cm)* and N_b are unknown and therefore, must be supplied before the equation is of practical value. These relations are determined by fitting to values derived by use of *Effuse* with *Mathematica* [5].

III.A Average distance traveled per particle, L

By fitting *Effuse* generated values for *L* with *Mathematica*, the following expression was found:

$$L(l,a) = l\big[-0.86\times\log_n(a) + 3.5505\big] + l^{2.31}\times 2.40.10^{-3} + 3.5a - 10 \tag{3}$$

where $\log_n a$ is the natural logarithm of the tube radius, a. This formula is valid over a range of tube dimensions: 5 cm ≤ l ≤ 100 cm and 0 cm ≤ a ≤ 2.5 cm.

III.B Average number of wall collisions, N_b

Analogously, an expression was found for the number of bounces, N_b, by fitting *Effuse* generated values for N_b with Mathematica [5]. The expression is given by

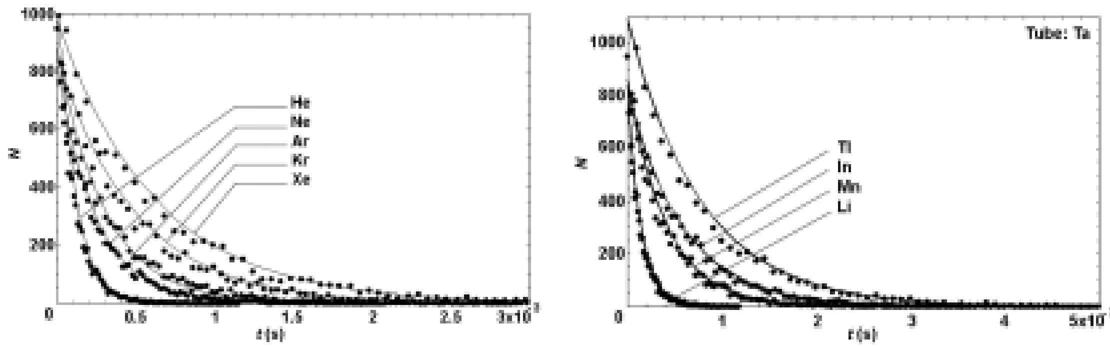
$$\mathrm{N_b} = \left[-0.1019 - \frac{0.103}{a}\right]\times l\times\log_n(a) + 0.4207\times l + 0.4253\times\frac{l}{a} + 0.4148\times \mathrm{a} + \frac{0.8}{a} - 0.195 + \left[11.85 + \frac{12}{a}\right]\times l^{2.31}\times 2.40.10^{-5} \tag{4}$$

Expression 4 is valid over a range of tube dimensions, 5 cm ≤ l ≤ 50 cm and 0 cm ≤ a ≤ 2.0 cm.

SIMULATION RESULTS

IV.A Stable elements

The formula has been thoroughly tested and found to hold for atoms or molecules with low or high enthalpies of adsorption, flowing through short or long tubes with small or large radii, as illustrated in Figure 1.

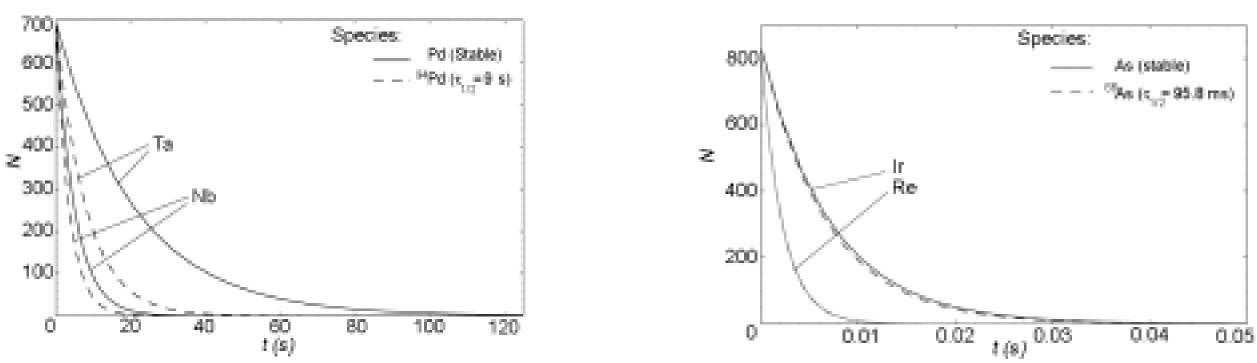


(a) **(b)**

Figure 1. Time distributions of (a) noble gases flowing through transport tubes made of any refractory metal (e.g., material Nb, Ta, Re and Ir); and (b) Li, Mn, In and Tl atoms flowing through tubes made of Ta as computed by use of Effuse (•) and Eq. 2 (—), at 2273 K. Tube length: 12.62 cm and tube radius: 0.293 cm.

IV.B Selected radioactive isotopes

The importance of selecting the most appropriate material for manufacture of the vapor transport system for selected radioactive isotopes is illustrated in Figure 2. As noted, Nb is a better choice for the transport of ^{94}Pd whereas either Ir or Re would be viable choices for the transport of ^{66}As. If the lifetimes of species are short with respect to their transport times, obviously, delay times attributable to adsorption/desorption processes can have dramatic influences on the rates at which radioactive species arrive at the ion source.



(a) **(b)**

Figure 2. Time distribution of (a) stable Pd and ^{94}Pd ($\tau_{1/2}$: 9 s) flowing through Nb and Ta tubes; and of (b) As and ^{66}As ($\tau_{1/2}$: 95.8 ms) flowing through Re and Ir tubes. Tube length: 12.62 cm and tube radius: 0.293 cm.

V. DISCUSSION

The utility of the formula lies in the fact that it can be used as a replacement for Monte-Carlo techniques to compute the time dependence for transport of a wide variety of elemental/molecular species through simple tubular systems on an absolute basis. Moreover, the equation permits separation of time dependence aspects of the diffusion-release and effusive-flow processes. Another utilitarian aspect of the equation is that it permits the choice of materials of tube construction that minimize the time required for transport of a given species through arbitrary geometry transport systems, on a relative basis.

[1] Alton, G. D., Beene, J. R., J. Phys. G: Nucl. Particle Phys., **24** (1998) 1347.
[2] Alton, G. D., and Liu, Y., Nucl. Instrum. and Meth. A, **438** (1999) 190.
[3] *Effuse* is a Monte-Carlo code, written by J. Dellwo and G. D. Alton, that is designed for calculating time dependent transport of gaseous elemental/molecular species through simple tubular systems.
[4] Present, R. D., *Kinetic Theory of Gases*, McGraw-Hill Book Company, (1958).
[5] Mathematica 3.0, S. Wolfram, Wolfram Research, Champaign, Il 61820, USA.