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

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Abstract. An analytical formula has been developed that accurately reproduces Monte Carlo simulations for the effusive-flow transport of chemically active species through tubular transport systems under ideal conditions, independent of species, tube material, and operational temperature. Through its use, the choice of materials of construction for a given transport system can be made on a relative basis that minimizes effusive-flow times, independent of 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 for noble gases as well as a variety of chemically active species.

INTRODUCTION

Excessively long delay times due to diffusion from solid or liquid target materials followed by their effusive-flow transport to the ion source are the principal means whereby short-lived radioactive species are lost between initial formation and acceleration at radioactive ion beam (RIB) facilities based on the ISOL technique [1]. Due to the low production rates and short lifetimes of species of research interest, RIB intensities are at a premium at such facilities and consequently, time delays due to each of these processes must be minimized. Since the residence times of chemically active species depend strongly on the enthalpies of adsorption of the species/surface combination as well as the operational temperatures used system during transport, it is desirable to choose low enthalpy of adsorption, highly refractory materials for constructing the transport system. We have developed an analytical formula for estimating transport times of species through tubular geometry transport systems. The results derived by its use can be used on a relative basis to select materials of construction that will minimize the transport times for any species for which enthalpies of adsorption are known, independent of size and geometry of the system. The equation accurately replicates results derived by use of Monte-Carlo statistical techniques.

Effusive Flow

In the case of a straight tube at constant temperature T and low pressure, transport through the system is referred to as free-molecular diffusion or Knudsen-flow. Under these conditions, a gas molecule collides many times with the walls of the tube before it encounters another molecule. Therefore, molecule-molecule collisions are negligible.

For an ideal gas in a tube of radius, a, and length, l, at low pressure, the steady-state flow rate, dN/dt, for particles of average velocity υ, flowing through the tube under a density gradient along the tube of dn/dz, is given by the following relation [2]:

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

where n is the particle density, υ is the Maxwellian velocity and k_B is Boltzmann's constant.

Time delays associated with adsorption that are excessively long in relation to the lifetime of the radioactive species can result in significant losses of

beam intensity in an ISOL facility [1]. Therefore, it is desirable to minimize residence times of atoms/molecules on surfaces in the target/ion source. The residence time, τ_{ad}, for a particle on a surface at temperature, T, is given by the Frenkel Equation (see, e.g., Ref [3]):

$$\tau_{ad} = \tau_0, \exp\{-H_{ad}/k_B T\} \qquad (2)$$

where H_{ad} is the heat of adsorption or enthalpy required to evaporate the atom or molecule from the surface, k_B is Boltzmann's constant, T is the absolute temperature and τ_0 is the time required for a single lattice vibration.

The desorption rate of atoms dN/dt in thermal equilibrium with a surface at temperature T, can be expressed by [4]:

$$\frac{dN}{dt} = P(T)A_0 Q_i k_B T.\exp\left[-H_{ad}/k_B T\right]\Big/\sigma N_0 h\Lambda^2 \qquad (3)$$

where $P(T)$ is the temperature-dependent probability that the particle will stick to the surface, Q_i is the internal partition function of the gas phase molecule, $\Lambda=(h^2/2\pi M k_B T)^{1/2}$ is the thermal wavelength, A_0 is the area of the surface having N_0 adsorption sites. Thus, the rate of desorption is governed by the enthalpy of adsorption, H_{ad}, and the temperature, T, of the surface.

Computer Simulations

The Monte-Carlo simulation program, *Effuse* [5], tracks particles by randomly choosing starting parameters (radial position and angle of injection) at entrance to the tube and starting angles at positions of de-sorption along the tube. The code accounts for residence times that particles spend on the walls of the transport system during passage. Knudsen-flow conditions are assumed, i.e., the mean free path, λ, is considered to be large with respect to the radius of the tube and therefore, collisions between particles are negligible. Consequently, each particle is assumed to travel in a straight line between collisions with the wall at temperature, T, at an average Maxwellian velocity, υ. After each particle is tracked through the system, the code records the average distance traveled per particle, L, and the average number of bounces, N_b, that the particle makes during transit through the tube. The code typically tracks 10,000 particles through a given system, starting at t=0s and ending when all particles have been transferred through the system.

Development of the Effusive-Flow Formula

The number of particles, N, radioactive particles with lifetime, $\tau_{1/2}$, remaining in a tube at time, t, after evacuation can be deduced by solving the time dependent form of Eq. 1, resulting in the following expression:

$$N = N_0 \exp\{-\lambda t\}\exp\{-t/\tau_c\} \qquad (4)$$

where N_0 is the number of particles in the volume at time $t=0s$, $\lambda = 0.693/\tau_{1/2}$; H_{ad} is the enthalpy of adsorption; τ_0 is a constant with value taken as $\tau_0 = 3.4.10^{-15}$s; L is the average distance that a particle travels during transit through the tube; and τ_C is the characteristic transport time given by

$$\tau_C = \tfrac{3}{4}\left[N_b \tau_0 \exp\{-H_{ad}/k_B T\} + L/\upsilon\right] \qquad (5)$$

The factor 3/4 was found by fitting Eq. 2 to Monte Carlo results derived for transport of identical particles through identical tubular systems under the same conditions.

Average distance traveled per particle, L

The average distance, L (cm), traveled by a particle during transit through a tubular system is assumed to be independent of tube material, species and temperature, T, and only a function of the length, l, and radius, a, of the tube. A formula for L was found by use of *Mathematica* [6] to fit to Monte Carlo results derived by use of *Effuse*. The resulting formula is given by:

$$\begin{aligned} L(l,a) = {} & l\left[-0.86\times\log_n(a) + 3.5505\right] \\ & + l^{2.31}\times 2.40.10^{-3} + 3.5a - 10 \end{aligned} \qquad (6)$$

where $\log_n(a)$ is the natural logarithm of the tube of radius a. Results derived by use of this formula are found to be in very good agreement with those obtained from *Effuse* for the following range of tube dimensions: 5 cm $\le l \le$ 100 cm and 0 cm $\le a \le$ 2.5 cm.

Number of wall collisions, N_b

A linear relation between the number of bounces, N_b, and the average distance traveled per particle during transit through a tubular system, $L(l,a)$, is found from *Effuse* calculations and after fitting with Mathematica and using the expression of $L(l,a)$, the expression of N_b can be written:

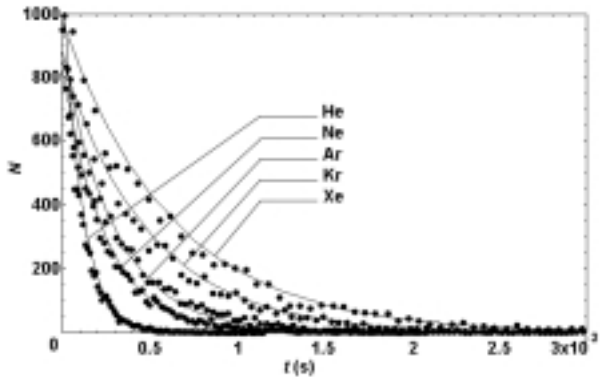


Figure 1. Time distribution of noble gas elements flowing through Nb, Ta, Re and Ir tubes as computed by use of Effuse (•) and Eq. 2 (—), at 2273 K. Tube length, l: 12.62 cm and tube radius, *a*: 0.293 cm.

$$N_b = \left[-0.1019 - \frac{0.103}{a}\right] \times l \times \log_n(a) + 0.4207 \times 1 + 0.4253 \times \frac{1}{a} + 0.4148 \times a + \frac{0.8}{a} - 0.195 + \left[11.85 + \frac{12}{a}\right] \times 1^{2.31} \times 2.40.10^{-5} \quad (7)$$

Again, results derived by use of this formula are found to be in excellent agreement with those of *Effuse* over the following range of tube dimensions: 5 cm ≤ l ≤ 50 cm and 0 cm ≤ a ≤ 2.0 cm.

Values of L and N_b, determined, respectively, from Eqs. 6 and 7, can then be used in Eq. 4, to calculate the number of particles, N, remaining in a tube of specified dimensions at time, t, for any atomic or molecular species for which enthalpies of adsorption, H_{ad}, are known.

Simulation Results

Noble gas elements

For these elements, residence times on the walls are negligible because of their very low enthalpies of adsorption, H_{ad}, and, consequently, the velocity dependent term, L/υ in Eq. 5 dominates. As noted in Fig. 1, the time dependent flow characteristics of these elements, derived by use of Eq. 5, are found to be in good agreement with *Effuse* values.

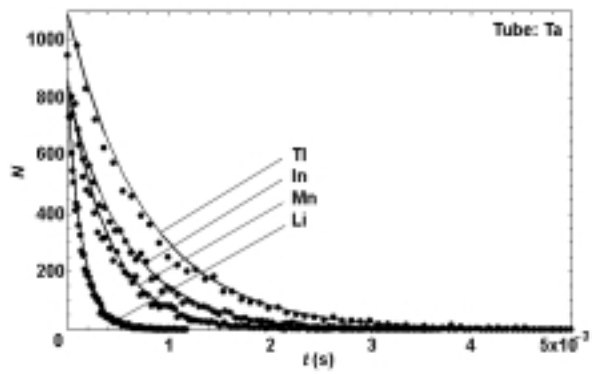


Figure 2. Time distribution of Li, Mn, In and Tl flowing through a transport tubes made of Ta as computed by use of Effuse (•) and Eq. 2 (—), at 2273 K. Tube length, *l*: 12.62 cm and tube radius, *a*: 0.293 cm.

Electropositive stable elements

Many of the less refractory and more electropositive elements move through a given transport system very quickly with characteristic times approaching those for noble gas elements of comparable mass when the transport tube is operated at high temperature. For these elements, the characteristic transit times are less sensitive to the materials of which the transport systems are made, as noted in Fig. 2 at the assumed operating temperature (i. e., T = 2273 K).

Electronegative stable elements

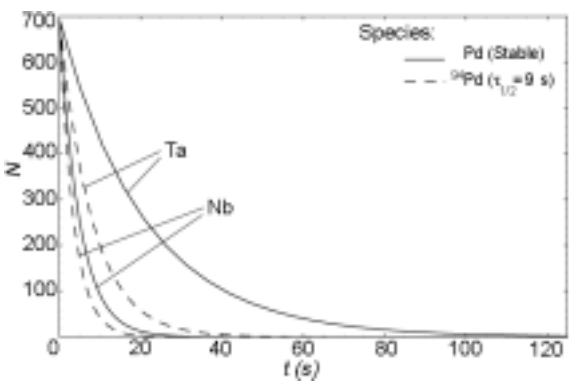
On the other hand, the characteristic times for more electronegative species are very sensitive to the choice of the materials of construction of the transport system and consequently vary widely. The transit times for these elements can be reasonably short, moderate or prohibitively long (see, e.g., Table 1).

Selected radioactive isotopes

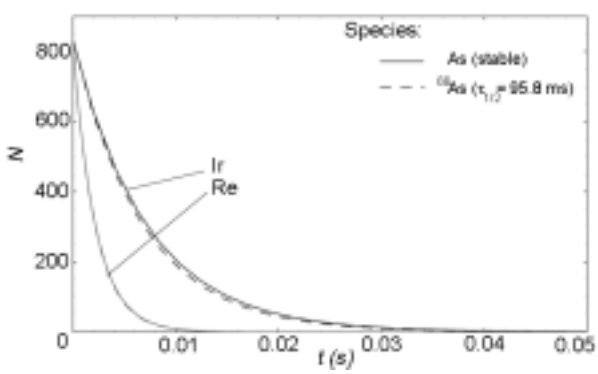
Delay times due to adsorption/desorption can have a dramatic influence on the rate at which radioactive particles arrive at the ion source if the species has a lifetime less than the characteristic transport time for the stable isotope of the element in question. The importance of selecting the most appropriate material for manufacture of the vapor transport system for selected radioactive isotopes in various tubes is illustrated in Figure 3. As noted in Figure 3, Nb is a better choice for the transport of ^{94}Pd whereas either Ir or Re would be a viable choice for the transport of ^{66}As.

Table 1. Characteristic effusive flow times, τ_C, of various electronegative elements through various tube materials. Tube length, *l:* 12.62 cm and tube radius, *a*: 0.293 cm. Temperature = 2273 K.

Characteristic effusive flow times, τ_c (s)				
Elements	Tube materials			
	Ta	Re	Ir	Nb
B	1070	60	118	454
C	10^9	10^6	10^6	10^9
N	10^9	421	143	10^9
O	10^{10}	81	10^{-4}	10^{10}
Si	1017	16	48	206
Ge	2	10^{-2}	10^{-1}	10^{-1}
Pd	21	10^{-3}	10^{-4}	5



(a)



(b)

Figure 3. Time distribution a) for stable Pd and ^{94}Pd ($\tau_{1/2}$=9s) flowing through Nb and Ta tubes and b) for As and ^{66}As ($\tau_{1/2}$=95.8ms) flowing through Re and Ir tubes. Tube length, *l:* 12.62 cm and tube radius, *a:* 0.293 cm.

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 under ideal conditions, 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.

ACKNOWLEDGEMENTS

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5. *Effuse* is a Monte-Carlo code, written by J. Dellwo and G.D. Alton, developed for calculating the time dependent transport of gaseous elemental/molecular species through simple tubular systems.
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