



Short communication

Gas-surface interfacial and gas-phase resistances to vaporizing cesium hydroxide and chloride in air



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ABSTRACT

The interfacial and the pure gas-phase resistances in air to vaporizing cesium species were evaluated in a comparison of the experimental vaporization flux and the maximum vaporization flux as calculated based on the kinetic theory of gas and the film theory. Gas-phase mass transfer resistances did not show great changes with the temperature, but their values were much greater than those associated with the interfacial resistances. The values of gas-phase resistances were at least a hundred times greater than interfacial resistances for the vaporizing cesium species investigated in this study.

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The vaporization rates of a metal species under vacuum condition can be estimated by its equilibrium vapor pressures [1]. However, the vaporization rates in a foreign gas cannot be estimated with the equilibrium vapor pressures due to the gas-phase resistance, which limits the transfer of vaporizing substances into foreign gas [2]. This study attempts to evaluate the resistances in a series and how they limit the transport of semi-volatile metal species in air.

The theoretical concept of the mass transfer resistance to vaporizing species by applying the classical kinetic theory of gas in conjunction with the film theory is shown in Fig. 1 [2,3]. Under an equilibrium condition, the vapor pressure at a liquid–gas interface, P_i , is equal to the vapor pressure at a liquid surface, P_s . The rate of collision of the gas molecules with the surface is readily calculated from the classical kinetic theory of gases. The colliding mole flux with a liquid surface is given by equation (1) [4,5].

$$J_v = P_i / (2\pi MRT)^{0.5} \quad (1)$$

In equation (1), J_v is the mole flux of gas molecules colliding with the liquid surface in $\text{mol m}^{-2} \text{s}^{-1}$, M is the molecular weight in g mol^{-1} , R is the ideal gas constant ($8.314 \times 10^{-5} \text{ m}^3 \text{ bar mol}^{-1} \text{ K}^{-1}$)

and T is the absolute temperature of gas in K. Some fraction α of these colliding molecules remains in the liquid while the fraction $1-\alpha$ rebounds to the gas. Under an equilibrium condition, vaporization must occur at the same rate of condensing molecules. Therefore, the net vaporization flux J_v is given by the difference between the vaporization and condensation rates of molecules at the liquid surface considering that T_s is equal to T_i as T .

$$J_v = 44.3\alpha(P_s - P_i) / (MT_s)^{0.5} \quad (2)$$

In equation (2) α is the condensation coefficient, and α and P_s are functions of only the liquid surface at the temperature T_s [3]. Under the equilibrium condition, P_i equals P_s , and there is no net mass transfer between the condensed phase and the gas phase. Under a non-equilibrium condition in which vaporization occurs, P_i is smaller than P_s because the gas molecules at the interface disappear into the surrounding gas. Under such a condition in which gas-phase resistance exists in the presence of a foreign gas, one can evaluate this gas-phase resistance with the film theory. The vaporization flux equations are analogous to Ohm's law, and the ratio of the driving force (the vapor pressure difference in this study) to the vaporization flux represents the resistance. The steady-state flux equations are given as equation (3) with the interfacial, gas-phase and overall mass transfer coefficients, k_i , k_g and K_g , respectively.

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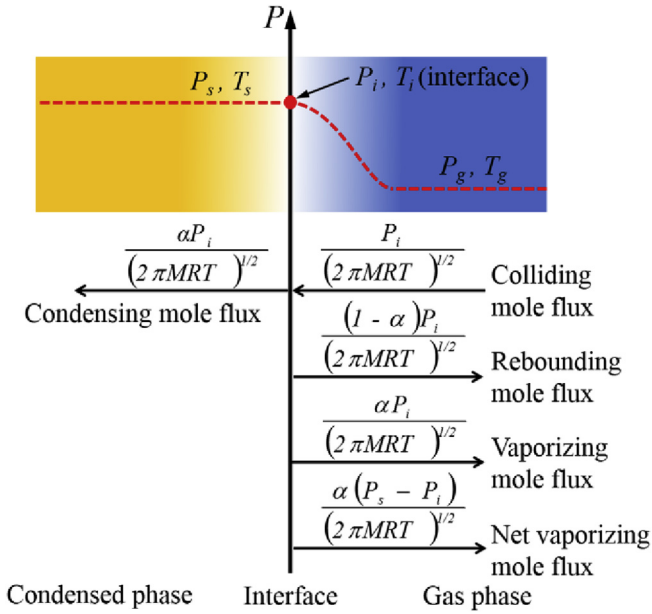


Fig. 1. Theoretical concept of the mass transfer resistance to vaporizing species.

$$J_v = k_i (P_s - P_i) = k_g (P_i - P_g) = K_g (P_s - P_g) \quad (3)$$

P_g in equation (3) is the partial pressure of a vaporizing substance in the surrounding gas. Combining equations (2) and (3), the rate of phase transition and the rate of transport into the foreign gas are determined by equation (4).

$$k_g(P_i - P_g) = 44.3\alpha (P_s - P_i)/(MT_s)^{0.5} \quad (4)$$

Solving equation (4) for P_i and substituting it into equation (3) gives equation (5).

$$J_v = k_g (P_s - P_g) / [1 + (MT)^{0.5} k_g / (44.3\alpha)] \quad (5)$$

The gas-phase resistance $1/k_g$ is obtained from the interfacial resistance $1/k_i$ and the overall mass transfer resistance $1/K_g$.

$$1/k_i = 1/K_g - 1/k_g = 0.0226(MT)^{0.5} / \alpha \quad (6)$$

The interfacial and pure gas-phase resistances in air to vaporizing metal species were evaluated in a comparison of the experimental vaporization flux and the maximum vaporization flux. The metal species investigated here were cesium species, as this species is the most important semi-volatile radioactive species emitted by the nuclear industry under normal as well as accidental operations [6–11].

A schematic diagram of the thermogravimetric analysis (TGA) system used in this study is shown in Fig. 2. This custom-made TGA system with a maximum weighing capacity of 10-g mainly consists of a TGA furnace, a control and data acquisition system, and a gas feeding system. A cylindrical crucible made of nickel alloy (Inconel 600) with a radius of 10.5-mm and a height of 20-mm was used to avoid an alkaline attack when using the alumina crucible. The investigated cesium species were hydroxide (CsOH) and chloride (CsCl). All chemicals used in study were samples with high purity levels (>99%), and all were supplied by Sigma Aldrich Chem. Co.. Isothermal TGAs for each cesium species were conducted while feeding compressed air into the TGA furnace at a rate of 1 L min⁻¹ and the exhaust gas was scrubbed with multiple metal-vapor

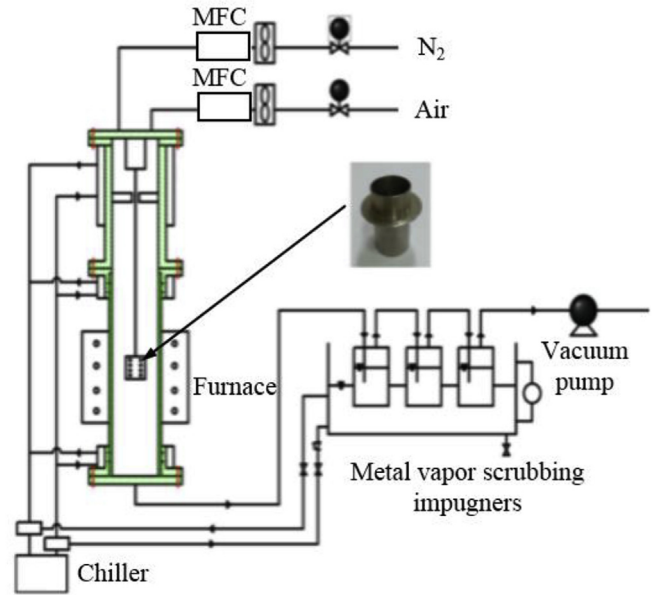


Fig. 2. Schematic diagram of the TGA system used in this study.

scrubbing impingers filled with a 5% HNO₃/10% H₂O₂ solution, as shown in Fig. 2. Each test was conducted by heating the furnace at a rate of 20 K min⁻¹ from room temperature to the investigated isothermal temperature, and then sustained at that temperature.

Assuming that the interfacial area for the vaporization of melted samples is constant and equal to the inner cross-sectional area of the cylindrical crucible, the vaporization flux can be obtained from the weight loss per unit of time. This is determined by equation (7).

$$J_v = - (AM)^{-1} (dW/dt) \quad (7)$$

In equation (7), A (m²) is the cross-sectional area of the crucible, M (g mole⁻¹) is the molecular weight of the sample cesium species and dW (g) is the weight change during the differential time, dt (s). The results of the isothermal TGAs for CsOH and CsCl at seven different isothermal temperatures are shown in Fig. 3. If the vaporization flux does not vary with time, the weight loss plots should show a linear relationship with time according to equation (7). All weight loss plots showed a nearly perfect linear relationship. Linear regression was done from 0% to 80% of the initial sample mass; these results are also shown in Fig. 3. The slopes of the fitted line shown in Fig. 3 reflect dW/dt in equation (7).

Under such a condition, in which the gas molecules at the interface are completely removed without the resistance of any foreign gas in an absolute vacuum, the maximum vaporization flux $J_{v,max}$ is determined by introducing P_i and setting it to zero in equation (2).

$$J_{v,max} = 44.3 \alpha (TM)^{-1/2} P_s \quad (8)$$

This maximum rate is very large compared with the mass-transfer rates encountered in most industrial equipment. The practical application of equation (8) requires the values of the condensation coefficient α (also called the sticking or accommodation coefficient). There is no useful theory to employ when predicting α , and there is no easy means of experimentally measuring it. Published α values for metal species range from 1.0 to 0.27 [12]. The α value of liquid potassium at 339.7–392.3 K was 0.95 ± 1 and that of potassium chlorides at 1126 K was 0.4 ± 0.05. The α value of cesium bromide was 0.27 ± 1 at 785–830 K and that of cesium iodide was 0.36 ± 1 at 757–727 K α values of simple metals such as

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