

VAPOR PHASE DIFFUSION IN A TRANSPIRATION APPARATUS

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ABSTRACT

The process used in the transpiration method to measure vapor pressure can be used to create a purification method for used nuclear fuel with some changes. As the focus of a purification method is to encourage mass transfer, changes must be applied to the method of transpiration to encourage both diffusive and convective mass transfer. This paper outlines a process to model the diffusion process of the purification process to serve as a preliminary step to more complicated, future models.

NOMENCLATURE

r : distance measured orthogonally from the center of the cylinder (m)
 r_1 : the cylinder's radius (m)
 z : distance measured from the bottom of the cylinder to the top (m)
 L : the cylinder's length (m)
 t : time (s)
 D : Diffusion coefficient (moles/(m² s))
 C_{sat} : Saturated vapor concentration (mol/m³)
 C_s : Steady-State Solution
 C_t : Transient Solution

INTRODUCTION

To make nuclear energy more economical and less hazardous, a variety of methods are being explored in the literature to recycle used nuclear fuel (UNF). Recycling UNF decreases the volume of waste to be stored and provides more efficient use of fissile material.

One method to recycle used nuclear fuel is based on the transpiration method which has traditionally been used to measure vapor pressure of low volatile compounds. The general principle of the transpiration method is to flow an inert or reactive gas over a heated sample and measure the change in mass of the sample over time. The change in mass can be

correlated with the saturated vapor of the sample. For accurate measurements, the apparatus is designed to minimize diffusive mass transfer.

For a purification process for used nuclear fuel, we want to maximize convective and diffusive mass transfer. As a result, relationships derived and used to model mass transfer for typical transpiration studies are not usable in this application as they do not include diffusive mass transfer [1]

As a step towards modeling the purification process for UNF, I modeled the concentration profile of a compound over a heated sample contained in an enclosed cylinder.

METHOD

A simplified diagram of the apparatus can be viewed in Figure 1.

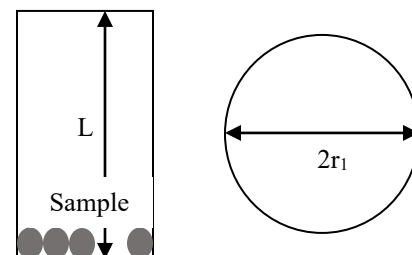


Figure 1 – Simple schematic of cylinder to be modeled.

As this is a preliminary step towards a more comprehensive model, the following assumptions have been made:

1. Mass transfer only occurs through diffusion.
2. The entire apparatus is isothermal – saturated pressure is the same everywhere.
3. The concentration is radially symmetric.
4. The vapor is monatomic – molecular weight is the same as the sample.

Applying these conditions provides the governing equation for the concentration profile [2]:

$$\frac{\delta C}{\delta t} = D \left[\frac{1}{r} \frac{\delta}{\delta r} \left(r \frac{\delta C}{\delta r} \right) + \frac{\delta^2 C}{\delta z^2} \right]$$

The initial condition is for r are

$$C(r, z, 0) = 0 \quad (1)$$

The boundary conditions for the r dimension are

$$C(0, z, t) < \infty \quad (2)$$

$$\frac{\delta C}{\delta r}_{r=r_1} = 0 \quad (3)$$

And for the z dimension are

$$C(r, 0, t) = C_{sat} \quad (4)$$

$$\frac{\delta C}{\delta z}_{z=L} = 0 \quad (5)$$

The full solution was obtained by solving for a steady-state solution and the transient solution by the method of separation of variables. The following steady-state solution was obtained:

$$C_s = \sum_{n=1}^{\infty} a_n Z_n R_n \quad (6)$$

where

$$a_n = \frac{2C_{sat}}{r_1 \lambda_n \cosh(\lambda_n L) J_1(\lambda_n r_1)} \quad (7)$$

$$R_n = J_0(\lambda_n r) \quad (8)$$

$$Z_n = \cosh(\lambda_n (z - L)) \quad (9)$$

$$\lambda_n \text{ are roots of } J_1(\lambda_n r_1) \quad (10)$$

Then the transient solution was determined to be

$$C_t = \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} A_{mn} R_n Z_m T_{nm} \quad (11)$$

Where

$$A_{mn} = \frac{4 \int_0^L \int_0^{r_1} C_s R_n Z_m r dr dz}{L r_1^2 J_1^2(\lambda_n r_1)}$$

$$Z_m = \sin(\nu_m z)$$

$$T_{mn} = e^{-D(\lambda_n^2 + \nu_m^2)t}$$

$$\nu_m = (m + 1/2) \frac{\pi}{L}$$

R_n and λ_n are the same as defined previously.

The full solution is the sum of the steady-state and transient solutions:

$$C(r, z, t) = C_s(r, z) + C_t(r, z, t)$$

CONCLUSIONS

An expression was obtained for the diffusion of a saturated vapor through long cylinder. Further work is needed to ensure that the shown solution ensures that the steady-state solution results in saturated vapor throughout the entire cylinder.

FUTURE WORK

To better represent the physical system, the model needs to become increasingly complex. In particular, convective mass transfer due to forced flow through the cylinder must be included. In addition, in a real system, the cylinder will not be isothermal in the z dimension. This means that saturated concentration will change as a function of z which will results in deposition out of the vapor phase and a decrease in the vapor concentration.

ACKNOWLEDGMENTS

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REFERENCES

- [1] Kvande, H.; Wahlbeck, P. G. Theory for the Determination of Vapour Pressures by the Transpiration Method. *Acta Chemica Scandinavica* **1976**, 30 A, 297–300.
- [2] William M. Deen. *Analysis of Transport Phenomena*. 2nd ed. 2012.

