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Sublimating Paradichlorobenzene Cylinders Oriented Horizontally in a Natural Convection Environment

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Abstract

Solid paradichlorobenzene pellets were melted and cast into cylinders. Each cylinder was then suspended horizontally and allowed to sublimate in air. Data on mass loss versus time were collected, and a sublimation rate was determined. The cylinder diameters were: 1 in. (2.54 cm), 1.5 in. (3.81 cm), and 2 in. (5.08 cm). The length of all cylinders was 10 in. (25.4 cm). Results of calculations made indicate that the Schmidt number was constant. The Sherwood number ranged from 4.7 to 21, while Rayleigh numbers ranged 14 x 10^3 to 113 x 10^3. The objective of this study was to derive an empirical equation for determining the mass transfer coefficient of horizontally suspended paradichlorobenzene cylinders that sublimated in a natural convection environment. An equation relating Sherwood and Rayleigh numbers was derived. For cylinder diameters ranging from 1 in (2.54 cm) to 2 in (5.08 cm), the results of this study indicate that the equation Sh = 2 $\times 10^{-4} \cdot$Ra + 2.385 correlates the data well, with $R^2 = 0.9999$.

Keywords: *heat transfer; mass transfer; sublimation; paradichlorobenzene.*

Nomenclature

C'	species concentration
A	surface area
C	concentration of Paradichlorobenzene
D	characteristic dimension (diameter)
D_{AB}	binary diffusion coefficient
g	acceleration of gravity
Gr	Grashof number = $\rho_{air} g D^3 (\rho_{v,s} - \rho_{v,\infty})/\mu^2$
h_m	overall mass transfer coefficient
k	thermal conductivity
M_p	molecular mass of paradichlorobenzene
M_a	molecular mass of air
$\dot{m}$	sublimation rate
N_A	molar flux
p_v	partial pressure of paradichlorobenzene
Ra	Rayleigh number = Gr·Sc
Re	Reynolds number = VD/ν
Sc	Schmidt number = ν/D_{AB}
Sh	Sherwood number = $h_m D/D_{AB}$
T	Temperature
T_m	mean or average temperature
T_w	temperature of cylinder surface
V	velocity

Greek symbols

β	volumetric thermal expansion coefficient
μ	viscosity of air
ν	kinematic viscosity
ρ	density of paradichlorobenzene vapor or of air

1. Introduction

Experiments have been performed in recent years to determine the sublimation rates for a variety of geometries. Nassif [1] performed experiments with a flat aluminum plate that had a machined recess on its upper surface. The upper surface was filled with molten naphthalene, which then

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solidified. The plate was placed into an open loop wind tunnel such that the upper surface was aligned with the air flow direction. The wind tunnel was then operated over a corresponding Reynolds number (based on plate length) that ranged from 40 000 to 2 000 000. The flow was turbulent over most of the upper surface of the plate. The plate was weighed before and after a finite amount of time to determine the sublimation rate of the naphthalene. The sublimation rate was then correlated with the Reynolds number. An empirical relationship was developed. The heat-mass transfer analogy was then used to compare the mass transfer results to heat transfer studies for flow over a flat plate, with excellent agreement.

Fite et al [2] described experiments performed with three different diameter cylinders [1 inch (2.54 cm), 2 inch (3.81 cm), and 3 inch (7.62 cm)] made of naphthalene, and paradichlorobenzene. These were cast and allowed to sublimate in an open loop wind tunnel, and situated with axes perpendicular to the flow direction. The flow velocities correspond to Reynolds numbers (based on cylinder diameter) that ranged 4 700 to 141 000. A correlation was derived to relate the Sherwood number to the Reynolds number. Results correlated well with existing heat transfer results using the heat-mass transfer analogy. The average uncertainty was calculated to be ± 9.7%.

Janna and Bruce [3] made paradichlorobenzene castings of cylinders, which were placed in an open loop wind tunnel. The diameters used were 1 in (2.54 cm), 2 inch (3.81 cm), and 3 inch (7.62 cm), and each was 18 inches (45.72 cm) in length. Air velocities ranged from 9.5 ft/s (2.9 m/s) to 106 ft/s (32.3 m/s) during the tests. A correlation was determined to relate the Sherwood and Reynolds numbers. The Reynolds numbers ranged from 6.19×10^3 to 1.60×10^5, with a Schmidt number of 2.23. The average uncertainty for the Sherwood number was + 4.6 to – 5.1 %. The average uncertainty for the Reynolds number was + 3.9 to – 3.5 %.

Sublimation rates of naphthalene casts of electronic components were measured by Schmidt [4]. Castings were subjected to natural and forced convection environments. The heat-mass transfer analogy was then used to determine the cooling requirements of electronic equipment.

A correlation between Sherwood and Rayleigh numbers for sublimating naphthalene was obtained by Goldstein [5]. Goldstein performed experiments in a fully insulated isolation room, using circular, square and rectangular geometries.

Bandrowski and Rybski [6] conducted naphthalene sublimation experiments using two different orientations of the same geometry. Theoretical models were advanced using the integral method for one case and a boundary layer approach for the other. It was found in the boundary layer model that increasing Schmidt numbers were accompanied by slight increases in the concentration of naphthalene vapor in the boundary layer, and a small increase in velocity. Bandrowski and Rybski found that the Sherwood number was proportional to the 1/5th power of the Grashof-Schmidt number product.

Sparrow and Niethammer [7] performed naphthalene sublimation studies to determine the effects of humidity on sublimation rates. It was concluded that humidity levels have no effect on the mass transfer rate of naphthalene over the temperature range studied.

Sogin [9] conducted inspirational studies of napthalene and paradichlorobenzene sublimation rates. Four-inch diameter disks of both substances were cast in steel rings, and placed on a glass surface. Rear faces of the plates were covered with paraffin, and the edges were taped. Only the forward faces of the plates were exposed and allowed to sublimate. Casts were placed in a wind tunnel for 5 to 180 minutes. Results were correlated using the heat-mass transfer analogy. The properties of paradichlorobenzene used in the Sogin study are used in the present work, including equations for partial pressure and density.

Snapp and Janna [8] conducted experiments with cast paradichlorobenzene cylinders, which were oriented vertically and allowed to sublimate in air. Sublimation rates were obtained in an effort to derive an equation relating the Sherwood and Rayleigh numbers.

In the present study, paradichlorobenzene cylinders were cast and suspended horizontally in air (as an extension of the Snapp and Janna study [8]). Cylinder diameters used were 1 in (2.54 cm), 1.5 inch (3.81 cm) and 2 inch (5.08 cm). The mass transfer rates will then be used to calculate Sherwood and Rayleigh numbers for each cylinder. Percent uncertainty will also be calculated so that the heat-mass transfer analogy may be studied further.

2. Experimental Procedure

Melted paradichlorobenzene granules were allowed to solidify in a mold to produce paradichlorbenzene cylinders. Prior to obtaining mass transfer data, tests were conducted on the cylinders to determine how much contraction took place during the solidification process. Graduated beakers were used to perform this test. Molten paradichlorobenzene was poured into one beaker, which was immediately covered while the paradichlorobenzene cooled to room temperature. Measurements were taken before and after the solidification process. The second test was performed in the same way but in this case, the beaker was left uncovered. After one hour had elapsed, the test specimens had cooled sufficiently, and the volumetric contraction of both samples compared favorably.

For the mass transfer portion of this study, high purity (99.8%) paradichlorobenzene was used for the castings. The paradichlorobenzene purchased was in the form of small crystals that required melting in order to cast a solid cylinder.

The molds were made of aluminum, and manufactured for heat transfer experiments. The diameters used are one, one and a half, and two inches. The molds were machined in a two pieces, both of which are held together with six bolts and a removable end cap. The specimens were easily removed once the paradichlorobenzene solidified, and cooled to room temperature (4 hours). Figure 1 is a photograph of two of the molds.

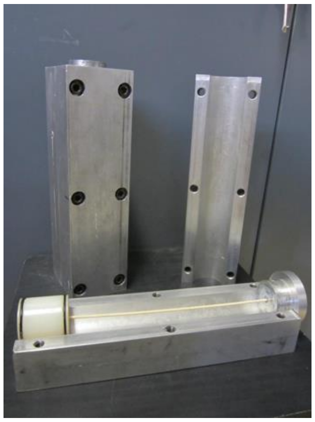

Figure 1. Molds used in the study

The molds were disassembled and thoroughly cleaned with alcohol prior to casting the cylinders. A small paraffin plug was made by pouring a small amount of hot wax into the bottom of the mold and allowing the wax to cool. The paraffin plugs were needed to prevent mass losses from the ends of the cylinders.

Paradichlorobenzene was then melted in a stainless steel pan placed on a hot plate. The melting temperature was roughly 170°F (76.7°C). The liquid was then poured through a funnel into one of the molds. A small amount of melted paradichlorobenzene was kept nearby, and added to the cylinder after volumetric contraction. After solidification, a paraffin plug was added to the top of the mold onto the cylinder to prevent unwanted losses. Temperature of each cylinder during the solidification process was nearly equal to room temperature, after approximately four hours, depending on the cylinder diameter.

The cylinders were carefully inspected for inclusions after taken from the molds. A number of the test samples had to be re-cast due to surface defects. Experience has shown that the liquid would flash cool on the sides of the mold when it was poured into a cold mold. Flash cooling effects were not desirable, and to alleviate this problem, the molds were heated slightly just prior to pouring the molting liquid into them. The sample quality was greatly increased because the rate of cooling (solidification) was slowed down considerably. Any remaining paradichlorobenzene in the stainless pan that used for melting was discarded after the volumetric contraction was accounted for.

The mass transfer tests conducted here lasted for several days in a laboratory environment. It took fifteen to twenty seconds from removal of the sample from the mold, to placement of the sample on the scale for measuring mass loss. This transfer time was considered insignificant compared to the overall time needed to complete the test. Because it was determined that the transient losses were insignificant, these losses were therefore not determined.

An Ohaus Adventurer electronic scale with a computer was used to obtain sublimation mass transfer data. The Ohaus Adventurer scale transmitted mass data through a RS232 port to the laptop. Prior to conducting the experiments, the scale was calibrated for accuracy by an independent laboratory. The reported accuracy was ±0.02 grams (0.00004 lbm), and it provided a digital readout in grams to two decimal places. The scale was set to transmit stable changes in mass of 0.05 gram (0.0001 lbm) increments. A Visual Basic program was written to record the change in mass, the time, the date, and trial number. The scale used had a weigh-below hook, which was used for this experiment. During the casting of the paradichlorobenzene cylinders, stainless steel wire was left hanging out of the top of the mold. The wire was 7 inch (17.8 cm) long, of 0.021 inch (0.53 mm) diameter. The weight of the wire was accounted for while taring the scale prior to conducting the tests.

After the paradichlorobenzene solidified, it was allowed to cool to approximately room temperature. The sample was then removed from the mold and suspended under the scale using the weigh-below hook. Then the Visual Basic program was started. The experiment was completely automatic at this point except for periodic measurements of the cylinder diameter, which were made by hand. Figure 2 shows an apparatus used in [9] study to obtain data on sublimation of vertically oriented cylinders.

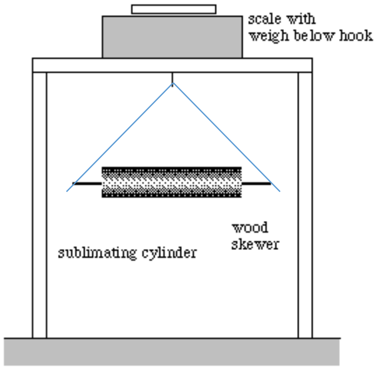

Figure 2. Paradichlorobenzene cylinder suspended horizontally under Electronic Scale [9]

The cylinder was allowed to sublimate until a deformation in the cylinder diameter was determined. A set of OD calipers were used periodically to measure the cylinder diameter at three different places. When the diameter of the cylinder at any location was reduced by more than 0.1 inches, the experiment was terminated. The results were then analyzed prior to casting another sample.

3. Mathematical Equations

In a heat transfer problem, natural convection density gradients are due to temperature differences, and they result in a movement of the fluid. In the analogous mass transfer problem, a concentration difference causes the gradients. Fick's law in mass transfer expression is analogous to Fourier's law in heat transfer:

$$N_A = -D_{AB} \left.\frac{\partial C_A}{\partial x}\right|_{x=0} \tag{1}$$

where the concentration gradient is the mass loss at the surface of the object. The equation describing molar flux is

$$N_A = h_m(C_A - C_\infty) \tag{2}$$

By equating these expressions, we obtain an expression for the convective mass transfer coefficient

$$h_m = \frac{-D_{AB} \left.\frac{\partial C_A}{\partial x}\right|_{x=0}}{(C_A - C_\infty)} \tag{3}$$

The volumetric thermal-expansion coefficient β relating density change to temperature differential is given by

$$\beta = -\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right) \tag{4}$$

In a difference form, we write Equation 4 as

$$\rho = \rho_m - \beta\rho_m (T - T_m) \tag{5}$$

Rearranging Equation 2 and substituting, we obtain

$$h_m = \dot{m} /[\, A_s(\rho_{v,s} - \rho_{v,\infty})] \tag{6}$$

At the surface of the cylinder, the paradichlorobenzene vapor density is $\rho_{v,s}$ and it is assumed to behave as an ideal gas. It may be calculated with the ideal gas equation. The vapor pressure of the paradichlorobenzene is needed to make this calculation, and the results given by Sogin [8] are used here. The Sogin equation for vapor pressure is given by

$$p_{vw} = 10^{(B1 - B2/Tw)} = 10^{(11.518 - 5\,946/Tw)} \tag{7}$$

where B_1 and B_2 are constants, and T_w is the temperature at the cylinder surface in °R. Equation 7 gives the vapor pressure in psf, which is converted to Pa after multiplying by 47.88. Vapor pressure data may also be found in [7] which when plotted, gives a straight line whose equation is

$$p_{vw} = 36.834(T_w) - 684.58 \tag{8a}$$

with a correlation coefficient of $R^2 = 1$. Temperature in the preceding equation is in °C, and pressure in Pa. If results from calculations made with the Sogin [8] relation is included, Equation 8a changes slightly to

$$p_{vw} = 37.756(T_w) - 745.14 \tag{8b}$$

with a correlation coefficient of $R^2 = 0.99$.

When vapor pressure of the paradichlorobenzene is determined, the vapor density is found with the ideal gas equation

$$\rho_{v,w} = \frac{p_{vw}}{R_v T_w} \tag{9}$$

The sublimation rate was found experimentally, and is given by,

$$\dot{m} = \frac{dm}{dt} \tag{10}$$

A dimensional analysis indicates that the data of this study may be correlated with the Sherwood, Grashof, Schmidt, and Rayleigh numbers, defined respectively as

$$\mathrm{Sh} = \frac{h_m D}{D_{AB}} \tag{11}$$

$$\mathrm{Gr} = \frac{D^3 g(C_{v,s} - C_{v,\infty})}{\mu^2} = \frac{\rho_{air} D^3 g(\rho_{v,s} - \rho_{v,\infty})}{\mu^2} \tag{12}$$

$$\mathrm{Sc} = \frac{\nu}{D_{AB}} \tag{13}$$

$$\mathrm{Ra} = \mathrm{Gr}\cdot\mathrm{Sc} = \frac{\rho_{air} D^3 g(\rho_{v,s} - \rho_{v,\infty})}{\mu^2} \cdot \frac{\nu}{D_{AB}} \tag{14}$$

The Grashof number represents the ratio of buoyancy to viscous forces; the Schmidt number is the ratio of momentum and mass diffusivities; the product of the Grashof and Schmidt numbers is the Rayleigh number, which is the ratios of buoyancy to viscous forces and momentum to mass diffusivities.

Several assumptions are necessary to solve the problem posed in this study:

- The density of the paradichlorobenzene is zero at a significant distance from the cylinder ($\rho_{v,\infty} = C_{v,\infty} = 0$). The facility used is over 160,000 cubic feet, and has unrestricted ventilation.

- There is incompressible isothermal flow where the concentration boundary layer starts at the surface of the cylinder.
- Chemical reactions do not take place.
- The physical properties of paradichlorobenzene vapor are constant and assumed to act like a perfect gas.

4. Results and Discussion

For all three cylinders, the raw data were graphed with cylinder mass on the vertical axis and time on the horizontal axis. Mass transfer rates were calculated using a linear curve fit. It was observed that the mass of the sublimating cylinders varied linearly with time, and so the assumption that the sublimation rate is constant is valid. Slight fluctuations in the mass versus time graphs were observed, and these were attributed to air movements in the lab. These fluctuations were minor, and assumed to not affect the results. The data obtained were analyzed to allow calculations of Sherwood and Rayleigh numbers.

During this study, the trial was stopped when the difference in the diameter, as measured vertically and horizontally, was 0.5%. This prevented the change in shape of the cylinder from affecting the results obtained. After the testing was complete, the cylinders were moved to a different part of the lab and allowed to sublimate for several more days. As the process continued, the diameter was noticeably larger when measured vertically than when measured horizontally.

The constants used in making the calculations are as follows

Table 1. Constants used in calculations

Ambient temperature	T_w = 79°F = 539°R	p_v = 236.5 Pa	(Eq 8b)
Gas constant	R_v = 56.55 J/(kg·K)	ρ_v = 0.009964 kg/m^3	(Eq 9)
Cylinder length	L = 0.254 m (= 10 in)	ρ_{air} = 1.265 kg/m^3	
Diffusion coefficient [8]	D_{AB} = 4.13 x 10^{-06} m^2/s	ν_{air} = 1.41x 10^{-5} m^2/s	
Schmidt number [8]	Sc = 2.23	μ_{air}= 1.79 x 10^5 N·s/m^2	

Table 2. Reduced data of this experiment

	1 inch	1.5 inch	2 inch
Sublimation rate $\dot{m}$ (kg/s)	1.53 x 10^{-7}	3.40 x10^{-7}	6.90 x10^{-7}
Diameter (m)	0.025 4	0.038 1	0.050 8
Surface area (m^2)	0.020 27	0.030 40	0.040 54
h_{m} (m/s)	0.000 757	0.001 122	0.001 708
Sherwood number	4.7	10.4	21.0
Grashof number	6 353	21 443	50 828
Rayleigh number	14 169	47 819	113 348

For mass transfer correlations, Goldstein et al [5] recommend that the Rayleigh number is used (rather than the Grashof number) to correlate the data. The objective is to minimize the separate dependence of the Sherwood number on the Schmidt number. A summary of other calculations made is provided in Table 2. A graph of the Sherwood number versus Rayleigh number is provided in Figure 3.

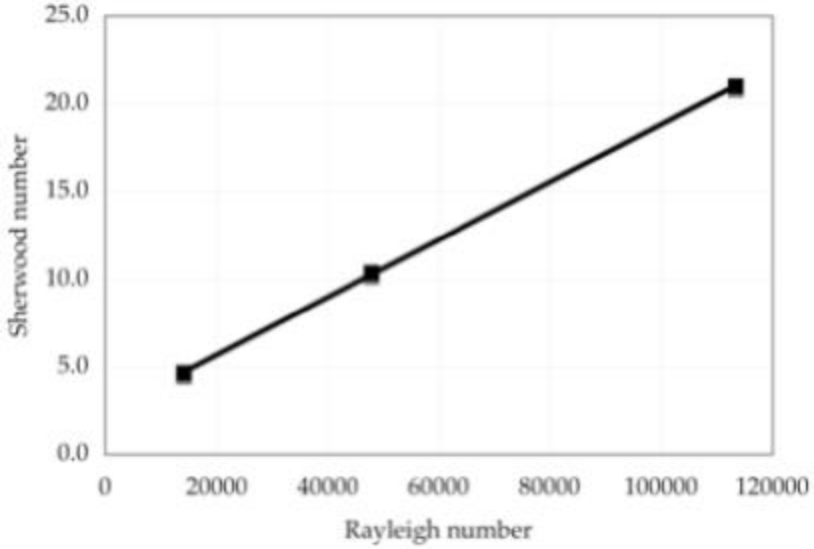

Figure 3. Sherwood Number versus Rayleigh Number for the data of this study (horizontally suspended cylinders)

For cylinder diameters ranging from 1 in (2.54 cm) to 2 in (5.08 cm), the results indicate that the equation

$$Sh = 2 \times 10^{-4} \cdot Ra + 2.385 \quad (15)$$

correlates the data well, with $R^2 = 0.9999$.

5. Error Analysis

There may have been slight variations in the paradichlorobenzene cylinder diameters during the mass transfer process, as well as slight manufacturing tolerances of the molds. These variations could have caused uncertainties in the results. Using the method outlined by Moffat [10], the errors in measurements made can be determined. Results are shown in Table 2. Because the Rayleigh number is the product of the Grashof and Schmidt numbers, the errors in the Grashof number will be the same as those of the Rayleigh number.

Table 3. Percent error of Sherwood and Grashof Numbers

Cylinder Diameter	Sh	Gr or Ra
1 inch (2.54 cm)	+0.9, -1	+3.04, -2.96
1.5 inch (3.81 cm)	+1, -1.1	+2.01, -1.99
2 inch (5.08 cm)	±1	+1.51, -1.49

Other sources of error may include slight uncontrollable temperature fluctuations, and air movements during the mass transfer process. The paradichlorobenzene cylinders were visually inspected during the tests, but it is possible that there may have been voids within the cylinder castings. Effects of humidity were shown by Sparrow and Niethammer [7] to have little effect on the sublimation of naphthalene. The effects of changes in atmospheric pressure were not investigated, but could have been a source of error due to the extended duration of the experiments.

6. Future Work

Using naphthalene rather than paradichlorobenzene and repeating these experiments could provide information on possible sources of error. Other geometries could be cast and studied using the procedures outlined here.

A comparison could be made between the results of this study and that of Snapp and Janna [9]. The dimensionless curve of this study (Figure 3 for horizontally suspended cylinders) can be compared to the Snapp curve for vertically suspended cylinders. The Rayleigh number in both curves span the range of approximately 0 to 120 000. The Sherwood numbers, however, show different ranges as expected. The two lines are not collinear, and the slopes are slightly different. Further analysis and more experimentation with another substance that sublimates in air (e.g., naphthalene) is recommended. The heat-mass transfer analogy can be studied as well, and results compared to existing heat transfer correlations. A plethora of work can still be performed in this area.

7. Conclusions

The results of this work show that there is a significant correlation between the average Sherwood and Rayleigh numbers for sublimating cylinders in a natural convection environment. The resultant correlation was found to be Equation (15). Figure 3 shows that the relationship between Rayleigh number and Sherwood number is linear. The square of the correlation coefficient for the line of best fit through the data is 0.99.

Because sublimation and heat transfer are similar mathematically, the traditional heat/mass transfer analogy can be investigated to relate published heat transfer results to those of this study. This broadens the application of the results of this experiment.

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Jeff G. Marchetta is an Associate Professor in Mechanical Engineering at the University of Memphis. He is a lifetime senior member of the American Institute of Aeronautics and Astronautics (AIAA) and a member of the American Society of Gravitational Space Research (ASGSR). He received the AIAA Abe Zerem Award for Distinguished Achievement in Research in 2000. He is the faculty advisor for the University of Memphis AIAA Student Branch. His research interests focus on the modeling of flows with free surfaces with applications in the management of fluids in reduced gravity, work sponsored by NASA and the Tennessee Space Grant Consortium.