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# The surface flow of water vapour

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Abstract. The flow rate of water vapour through glass parallel plate channels of widths of the order of  $1.5 \,\mu\text{m}$  has been measured in the region of surface flow. An equation for surface flow, which agrees with the experimental results, is derived for the case of multi-layer adsorbed films.

## 1. Introduction

The flow of vapours through narrow artificial channels and porous materials is usually taken as the sum of the gas phase flow and the surface flow.

In order to study surface flow quantitatively experiments have been carried out on the flow of water vapour through parallel plate glass channels where the channel dimensions are known. Therefore it has been possible to compare experimental results on surface flow directly with a theoretical model. Such a comparison was not previously possible since the effects of surface flow had only been observed in porous materials. A review of surface and gas phase flow in porous materials is given by Barrer (1967).

This paper describes a series of experiments carried out on the flow of water vapour through parallel plate glass channels of widths between  $1.21 \,\mu\text{m}$  and  $1.80 \,\mu\text{m}$ . The results are interpreted in the light of the theoretical equations which are derived to take account of surface flow.

#### 2. Theory

The rate of flow of vapour Q through a narrow channel is given by

$$Q = Q_{\rm a} + Q_{\rm g}$$

where  $Q_a$  is the rate of flow in the adsorbed phase (surface flow) and  $Q_g$  is the rate of flow in the gas phase.

An equation for  $Q_g$  has been derived by Roberts (1969, 1971), namely

$$Q_{g} = -\frac{h^{2}b}{2}\left(\frac{\pi M}{2RT}\right)^{1/2}f(\alpha)\frac{\mathrm{d}P}{\mathrm{d}x},$$

where b is the channel breadth, h is the channel width, M is the molecular weight of the gas or vapour, dP/dx is the pressure gradient in the channel and  $f(\alpha)$  is a function of  $\alpha$  (the ratio of the channel width to the mean free path) and the surface-gas combination.

The surface flow  $Q_a$  is dependent on the amount of adsorption and on the pressure gradient which exists within the adsorbed film. The experimental results which are

described in §3 indicate that surface flow only becomes appreciable when the ratio of the pressure in front of the channel  $P_f$  to the saturation vapour pressure  $P_0$  is greater than 0.94 which corresponds to an adsorbed film of thickness approximately 0.7 nm. Therefore for the purpose of designing a model for surface flow, only multi-layer films will be considered which have the same values of density  $\rho$  and viscosity  $\eta$  as the liquid phase.

The pressure within the film which may be used in the viscous flow equation is that defined by Derjaguin as the disjoining pressure  $P_{dis}$  (see Derjaguin 1934, Frenkel 1946). Using this parameter Churaev (1963) derived an equation for surface flow which when applied to a parallel plate channel becomes

$$Q_{\rm a} = -\frac{2b\rho S^3}{3\eta} \frac{\mathrm{d}P_{\rm dis}}{\mathrm{d}x}$$

where S is the film thickness. By assuming localized equilibrium between the film and the gas phase Churaev (1963) also showed that for a film on a flat surface

$$P_{\rm dis} = \frac{\rho R T}{M} \ln \left( \frac{P}{P_{\rm o}} \right)$$

where P is the pressure above the film. Hence by substituting in the previous equation

$$Q_{\rm a} = -\frac{2b\rho^2 RT}{3\eta M} \frac{S^3}{P} \frac{\mathrm{d}P}{\mathrm{d}x}$$

and therefore the total flow becomes

$$Q = K_1 \frac{S^3}{P} \frac{dP}{dx} + K_2 f(\alpha) \frac{dP}{dx}$$
(1)

where

$$K_1 = \frac{2b\rho^3 RT}{3\eta M}$$

and

$$K_2 = \frac{h^2 b}{2} \left(\frac{\pi M}{2RT}\right)^{1/2}.$$

In order that equation (1) may be used to interpret the experimental results the variation of film thickness with pressure is required. As far as it is known the only results available for multi-layer adsorption of water vapour on glass are those of Derjaguin and Zorin (1957) and Frazer (1929). The two sets of results are not in close agreement, but the general shape of the respective isotherms is the same. Due to the improved technique of Derjaguin and Zorin their results were used and the equation

$$S = \exp\left(\frac{\sigma P}{P_0} - \gamma\right) \tag{2}$$

was empirically evaluated for the adsorption of water vapour on glass at room temperature, where  $\sigma = 40.73$  and  $\gamma = 38.72$ .

Equation (1) can only be used directly if S is constant along the channel, therefore since S varies greatly with pressure in the region of surface flow, equation (1) is modified so that it may be compared with the experimental results. The experiments were carried

out under steady-state conditions, hence Q is constant along the channel and dQ/dx = 0. Therefore from equations (1) and (2)

$$\frac{\mathrm{d}Q}{\mathrm{d}x} = \left\{ \frac{K_1}{P} \exp\left(\frac{3\sigma P}{P_0} - 3\gamma\right) + K_2 f(\alpha) \right\} \frac{\mathrm{d}^2 P}{\mathrm{d}x^2} \\ + \left\{ K_1 \left(\frac{3\sigma}{PP_0} - \frac{1}{P^2}\right) \exp\left(\frac{3\sigma P}{P_0} - 3\gamma\right) + K_2 \frac{\mathrm{d}f(\alpha)}{\mathrm{d}P} \right\} \left(\frac{\mathrm{d}P}{\mathrm{d}x}\right)^2 = 0.$$

By re-arranging and assigning z = dP/dx the equation becomes

$$\frac{\mathrm{d}z}{\mathrm{d}x} = \frac{-\left\{K_1\left(\frac{3\sigma}{PP_0} - \frac{1}{P^2}\right)\exp\left(\frac{3\sigma P}{P_0} - 3\gamma\right) + K_2\frac{\mathrm{d}f(\alpha)}{\mathrm{d}P}\right\}z^2}{\left\{\frac{K_1}{P}\exp\left(\frac{3\sigma P}{P_0} - 3\gamma\right) + K_2f(\alpha)\right\}} = -f(P)z^2.$$

But

$$\frac{\mathrm{d}z}{\mathrm{d}x} = \frac{\mathrm{d}z}{\mathrm{d}P}z$$

therefore

$$\frac{\mathrm{d}z}{z} = -f(P)\,\mathrm{d}P.$$

The boundary conditions were assumed to be  $P = P_f$  and  $z = z_f$  at x = 0 and  $P = P_b$  at x = l. Therefore at any channel cross section z is given by

$$z = z_{\rm f} \exp F(P)$$

where

$$F(P) = \int_{P}^{P_{f}} f(P) \,\mathrm{d}P$$

By substituting back with z = dP/dx,  $z_f$  may be determined from the integral

$$z_{\rm f} \int_0^l \mathrm{d}x = z_{\rm f} l = \int_{P_{\rm f}}^{P_{\rm b}} \frac{\mathrm{d}P}{\exp F(P)}$$

The values of  $z_f$  and  $P_f$  are then substituted back into equation (1) and the value of Q determined. Both integrals and the substitution of  $z_f$  and  $P_f$  in equation (1) were carried out by a computer. The variation of Q with  $P_f$  for different values of  $\Delta P = P_f - P_b$  is shown in figures 1 and 2 for a channel of width 1.50 µm (figure 2 is a blown-up detail of figure 1). The values used of the various parameters which appear in the equation were l = 9.7 mm, b = 89.0 mm, M = 18,  $T = 24.0^{\circ}$ C,  $\rho = 10^3$  kg m<sup>-3</sup>,  $\eta = 0.01$  poise and  $P_0 = 22.35$  Torr.

From the results it may be shown that Q is directly proportional to b and inversely proportional to l. No simple relationship exists between Q and any of the other parameters in the region of surface flow.

An interesting feature of the flow is that of the variation of pressure along the channel for values of  $\Delta P$  of the order of 2 Torr. As shown in figure 3 as  $P_{\rm f}$  extends into the surface flow region but with  $P_{\rm b}$  outside the region, the pressure gradient varies enormously along the channel, being maximum at the low pressure end where surface flow is almost zero.



**Figure 1.** Variation of Q with  $P_t$  for  $h = 1.50 \,\mu\text{m}$ .  $\Delta P = 2 \,\text{Torr}, ---\Delta P = 1 \,\text{Torr}, ----\Delta P = 0.5 \,\text{Torr}.$ 



Figure 2. Variation of Q with  $P_f$  for  $h = 1.50 \,\mu\text{m}$ .  $\Delta P = 2 \,\text{Torr}, ---\Delta P = 1 \,\text{Torr}, ----\Delta P = 0.5 \,\text{Torr}, \text{experimental results are indicated by }, \blacksquare \text{ and } \blacktriangle \text{ respectively.}$ 

The variation of Q with  $P_{\rm f}$  for different values of h is shown in figure 4 where  $\Delta P = 1$ Torr.

#### 3. Experimental procedure and results

The experiments were an extension of those described by Roberts (1969, 1971) where experiments on the flow of gases between parallel plates are described. The equipment and procedure used in the present investigation were similar to those described by



Figure 3. Variation of  $P - P_b$  with x in the channel with  $\Delta P = 2$  Torr.  $-P_f = 20$  Torr,  $-P_f = 22$  Torr,  $-P_f = 22$  Torr.



Roberts (1971) except that the temperature control system had been improved so that variations in temperature during an experiment were less than  $\pm 0.05^{\circ}$ C.

The channel surfaces were kept as clean as possible by thorough cleaning before the setting up of the channel and by evacuating the whole system to a pressure less than  $10^{-7}$  Torr at the beginning of each set of experiments. The procedure for each experiment was such that the water vapour was allowed to flow under equilibrium conditions through the channel for at least 3 hours before commencing with the actual experiment.

This procedure ensured that there was no appreciable adsorption and desorption effect in the system.

The main sources of error in the experiments were the measurement of plate separation, which was less than 2% and the error due to variation in temperature which was usually less than 5%, but was as big as 20% in the case of  $P_f/P_0$  greater than 0.9. However, since the surface flow observed was much greater than 20% of the expected flow in the gas phase this large error could be tolerated. Unfortunately it was not possible to carry out experiments with  $P_f$  greater than approximately 21.5 Torr due to the effect of water vapour on the micromanometer pressure gauge (referred to in the earlier paper) as the saturation vapour pressure is approached.

Experiments were carried out with channels of widths 1.21 µm, 1.50 µm and 1.80 µm, and differential pressures between 0.7 Torr and 2.5 Torr. Some of the results are shown in figure 4 where Q is plotted against  $P_f$  for  $\Delta P = 1$  Torr. Since the differential pressure could not easily be set at a particular value, the values of Q have been adjusted to those corresponding to  $\Delta P = 1$  Torr by means of the theoretical results shown in figures 1 and 2. However, the actual differential pressures of the results shown in figure 2 were within the range 0.91 Torr to 1.10 Torr, therefore the amount of adjustment was less than 15% of the measured value of Q.

The results for  $h = 1.5 \,\mu\text{m}$  are also shown in figure 2 to demonstrate the variation of Q with  $\Delta P$ . The same adjustments for  $\Delta P$  were also used but in this case the actual values of  $\Delta P$  were within  $\pm 0.25$  Torr to the values quoted in the figure.

The experiments described so far were carried out with values of l = 9.7 mm and b = 89.0 mm. Other experiments were carried out with channel widths as close as possible to 1.50 µm but with different values of l and b, namely l = 6.31 mm, b = 92.3 mm and l = 9.4 mm, b = 43.8 mm. These experiments confirmed the theoretical prediction that Q is directly proportional to b and inversely proportional to l.

## 4. Discussion

The good agreement shown in figures 2 and 4 between the theoretical evaluation and the corresponding experimental results clearly illustrates the existence of surface flow in the case of multi-layer adsorption. It is still feasible that surface flow due to a fraction of a monolayer exists, but this could not be detected with the present experiments. A model for surface flow was proposed by Jones (1951) who assumed that the adsorbed film is an ideal two-dimensional gas. Agreement between this model and the experimental results was not possible, therefore it must be regarded as a model to be used with partially full monolayers.

The other models studied are only applicable to films of low surface concentration such as that proposed by Hill (1956) or involve numerical constants which cannot be theoretically evaluated. Hence these models are not easily compared with the experimental results.

The channels were produced from optically flat glass surfaces. A study of their surface topography with a scanning electron microscope showed that the surfaces were flat to within 20 nm but contained random island irregularities with dimensions up to 100 nm. However, if the irregularities do not possess sharp edges their effect on the flow should be small since they increase the values of b and l proportionally and b/l is therefore constant.

During the course of publication it was suggested that capillary condensation occurs near the entry to the channel. This is considered unlikely since the highest value of  $P_{\rm f}/P$  used was 0.963, which due to the Kelvin equation for capillary condensation corresponds to a cylindrical surface of radius 0.015 µm. Once the edges of the channel are occupied by condensate the radius of the curved surfaces along the channel edges is greater than or equal to half the channel width, which is a factor of 40 greater than the radius needed for capillary condensation.

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