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CAPILLARY RISE WITH MENISCUS EVAPORATION FOR Kn ≥ 0.01

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UDC 532.68

A meniscus step in a capillary has been observed at reduced gas pressures; a physical explanation is given.

Capillary impregnation is widely used in temperature-control systems (porous evaporative cooling) for use at various pressures and temperatures. Research has shown [1] that when an open-pore material is wetted at a certain external pressure, there is a sharp change in the liquid position, with the boundary descending, which affects the thermostatic control. The thicker the specimen and the smaller the capillary radius, the lower the pressure at which the step occurs.

We have examined impregnation accompanied by evaporation subject to changes in the thermodynamic parameters for the medium and the liquid. The apparatus and methods have been described in [2].

We used cylindrical glass capillaries under a glass vacuum chamber cover in the strictly vertical position. The menisci were observed and photographed via an optical system. The time was recorded by a timer and a cine camera with built-in clock. A capillary was fixed in a holder fitted with a measurement scale (division 4×10^{-4} m) and filled by bringing up a cell containing the liquid to the lower end. The liquids were outgassed distilled water, ethyl alcohol, benzene, acetone, dibutyl phthalate, and glycerol.

The equilibrium rise was measured in two ways for various gas pressures: 1) the capillary was filled at atmospheric pressure, after which the pressure was reduced; and 2) the liquid was introduced at reduced pressure, the pressure then being raised to atmospheric. The experiments were done with a single radius and various lengths or with various radii but a single length. The minimum length was equal to the maximum height of rise at atmospheric pressure for the given radius, while the maximum was 0.25 m. The radii varied from 0.17×10^{-3} to 0.50×10^{-3} m.

When the pressure was reduced from atmospheric to 1.33×10^3 Pa, the rise was unaltered and the menisci remained fixed in all the capillaries. At 1.20×10^3 Pa, with capillaries 0.25 m long (or on further pressure reduction for shorter capillaries), the menisci for water descended somewhat below the maximum rise in the atmosphere. At 0.67×10^3 Pa, the menisci for water in all the capillaries remained at certain heights characteristic of the radius and length, with no alteration as the gas pressure was reduced further. Figure 1 shows that the sinking was the larger the greater the length for a given radius. In capillaries with the minimum length, there was no change in menuscus position as the pressure was reduced. As the step $\Delta \ell$, we took the difference between the heights of the menisci at atmospheric pressure and at 0.67×10^3 Pa. Figure 2 shows that $\Delta \ell = 0$ for capillaries whose lengths were equal to the maximum rise in air. The value of $\Delta \ell$ increases with the length for r = const, while it decreases as the radius increases for L = const (curves 1-6). For $r = 0.17 \cdot 10^{-3}$ m or less, steps occurred even when the length was about 5 mm greater than the maximum rise for water at atmospheric pressure. For $r > 0.4 \cdot 10^{-3}$ m, and for all the lengths ($L \leq 0.25$ m), there was virtually no shift for water.

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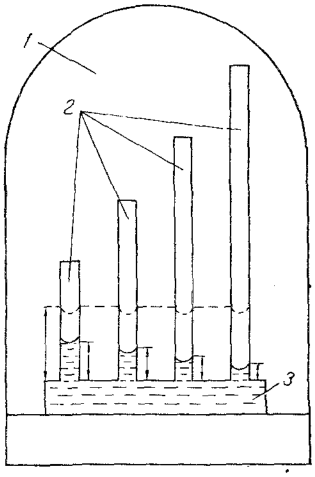

Fig. 1

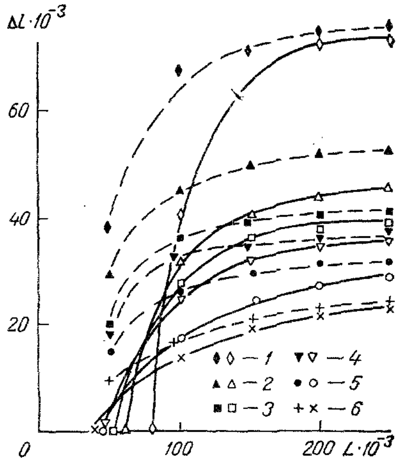

Fig. 2

Fig. 1. Menisci for water in capillaries ($r = 0.29\cdot10^{-3}$ m) with various lengths (from left to right 0.10; 0.15; 0.20; 0.25 m) when the gas pressure is reduced from atmospheric (dashed line) to 0.67×10^{3} Pa (solid lines): 1) vacuum chamber; 2) capillaries; 3) cell containing liquid.

Fig. 2. Meniscus step $\Delta\ell$ in m for water as a function of capillary length L in m for radii of $r = 0.17\cdot10^{-3}$ m (1); $0.23\cdot10^{-3}$ (2); $0.28\cdot10^{-3}$ (3); $0.3\cdot10^{-3}$ (4); $0.32\cdot10^{-3}$ (5); $0.34\cdot10^{-3}$ m (6); the dashed lines are the steps when the meniscus rises on account of increased gas pressure (first method), and the solid lines are for the meniscus falling with the pressure reduced (second method).

The precise sinking varies with the length and radius. With long capillaries ($r/L \lesssim 3\cdot10^{-3}$), the menisci descended stepwise, with heavily damped oscillations, while for $r/I > 3\cdot10^{-3}$, the sinking was comparatively slow and without oscillation.

If the liquid was brought up to a capillary at 0.67×10^{3} Pa (first method), the maximum rise for water was somewhat less than the height found on reducing the pressure from atmospheric to 0.67×10^{3} Pa. The hysteresis is due to differences in the wetting angle θ on descent (in a wetted capillary) and on rise (in a dry one). A similar pattern was observed when the pressure was raised starting from 0.67×10^{3} Pa to atmospheric (the water rose to a height a few mm less than when the capillaries were filled in the atmosphere, Fig. 2). Here $\Delta\ell$ (first method) was not zero in any of the capillaries, even ones whose lengths were less than the maximum rise in the atmosphere with water (curve 3 in Fig. 2). The solid and dashed lines in Fig. 2 show that the first method gives $\Delta\ell$ larger than the second. The rise on raising the gas pressure occurred stepwise but without oscillation.

The value of $\Delta\ell$ decreased linearly as the radius increased for a fixed length at 0.67×10^{3} Pa. The slopes of the curves for capillaries having $L \geq 0.10$ m were approximately the same but were reduced for $L = 0.05$ m. That length was taken as equal to the maximum rise for water in the atmosphere for the capillaries with the average radius ($r = 0.28\cdot10^{-3}$ m), so for $r < 0.28\cdot10^{-3}$ m that length was less than the maximum rise for water at atmospheric pressure, and the step was reduced in that case.

There is a marked change in the equilibrium meniscus position for water (step) on passage through the pressure range $\Delta P = (1.33–0.67)\cdot10^{3}$ Pa, the exact effect being dependent on the capillary radius and length.

Similar experiments were performed with the other liquids. If the liquid evaporates slowly (dibutyl phthalate or glycerol), there is no step on reducing the pressure to 0.13×10^{3} Pa, whereas for rapidly evaporating ones (water, alcohol, benzene, or acetone), the steps are prominent. The step for ethanol in capillaries having $r = 0.29\cdot10^{-3}$ m on reducing the pressure from atmospheric to $P = 2.00\cdot10^{3}$ Pa with $L = 0.1$ m was $3.2\cdot10^{-3}$ m, while for $L = 0.25$ m it was $\Delta\ell = 4.8\cdot10^{-3}$ m. For acetone, reduction from atmospheric pressure to 6.00

TABLE 1. Surface Temperatures for Various Liquid and Pressures at which Steps Occur

Liquid	P_{step} $\times 10^{-3}$,	T_ℓ in capillary, K	$P_s \cdot 10^{-3}$, Pa at T_ℓ	$P_s \cdot 10^{-3}$, Pa at $T=293$ K
Water H_2O	0,67	279—281	1,07	2,27
Alcohol C_6H_6O	2,00	279—280	2,67	6,00
Benzene C_6H_6	4,67	281	5,33	9,87
Acetone C_3H_6O	6,00	268	6,67	26,67

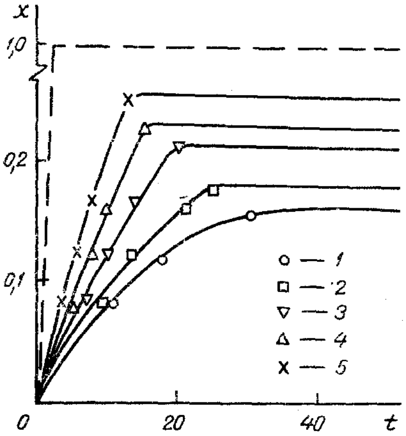

Fig. 3

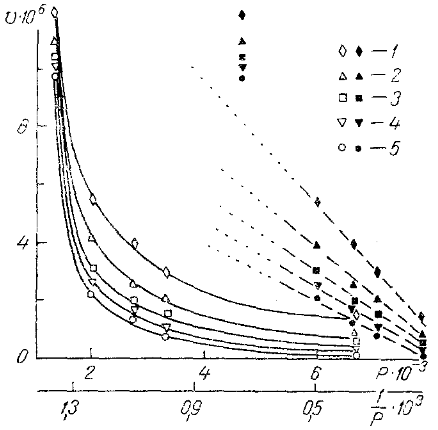

Fig. 4

Fig. 3. Entry of distilled water into a $r = 0.28 \cdot 10^{-3}$ m capillary at $p = 0.67 \cdot 10^3$ Pa (solid lines) and at atmospheric pressure (dashed line), capillary lengths 1) 0.25 m; 2) 0.20; 3) 0.15; 4) 0.10; 5) 0.07 m, x dimensionless, τ in sec.

Fig. 4. Rate of evaporation v for water in m/sec in a capillary having $r = 0.24 \cdot 10^{-3}$ m as a function of pressure P in Pa (solid lines) or 1/P in 1/Pa (dashed lines) for $\Delta h = 5 \cdot 10^{-3}$ m (1); $10 \cdot 10^{-3}$ (2); $15 \cdot 10^{-3}$ (3); $20 \cdot 10^{-3}$ (4); $25 \cdot 10^{-3}$ m (5).

$\times 10^3$ Pa gave $\Delta\ell$ of 3.0×10^{-3} and 4.3×10^{-3} m correspondingly in those cases.

The step pressure varies with the liquid but is always less than the saturation vapor pressure P_s at the working temperature (Table 1); the latter is less by several orders of magnitude than the pressures used for the slowly-evaporating liquids, e.g., $T_\ell = 298$ K $P_s = 0.25 \cdot 10^{-2}$ Pa for butyl phthalate, so those liquids showed no steps.

With water at 0.67×10^3 Pa, we found that the rate of rise was much less than at atmospheric pressure. The time needed to reach the equilibrium state for $r = (0.22–0.32) \cdot 10^{-3}$ m at atmospheric pressure was 1-2 sec, whereas at the lower pressure, the time was 10-20 times longer in the same capillaries. Figure 3 shows the relative rise ($x = \ell/\ell_\infty$) as a function of time for capillaries with various lengths, and at 0.67×10^3 Pa, that time increases with the length. Similar variations occurred with ethanol, benzene, and acetone.

One reason for the variation in rise rate and the step descent at pressures corresponding to $Kn = \Lambda/2r > 0.01$ is that the contact angle increases [2]. For example, with $r = 0.30 \cdot 10^{-3}$ m, the wetting angle for water varies from 18° at atmospheric pressure to 44° at 0.67×10^3 Pa. If one assumes that contact-angle change is the only cause of the step, the maximum rise in such a capillary, no matter what the length, should decrease from 47×10^{-3} to 35×10^{-3} m, and in that case $\Delta\ell = 12 \cdot 10^{-3}$ m, whereas we found that it was substantially larger than this (e.g., with L = 0.25 m, $\Delta\ell = (36–38) \cdot 10^{-3}$ m, Fig. 2) and was dependent on the length above the meniscus. The contact angle is therefore not the only reason for the descent at pressures characterized by $Kn > 0.01$.

A second possible cause is that there is a difference in the total pressure along the capillary for $0.01 < Kn < 0.1$ when the liquid evaporates. To examine the evaporation from capillaries, we used communicating U-shaped capillaries with bends having the same length of 0.25 m, which were filled to the top with water. The required pressure was set up, and the descent in the bends was used to measure the evaporation rate, where the error in determining the meniscus sinking rate was not more than 5%. The results were found to be highly reproducible for capillaries having the same radius.

Figure 4 shows the water evaporation rate as a function of pressure P or the reciprocal 1/P for given menuscus positions. Above 2.0×10^3 Pa, the points on v = f(1/P) lie on a straight line, i.e., in that P range, the evaporation rate is inversely proportional to the gas pressure, which is characteristic of a diffusion-limited process. Below that pressure, v = f(1/P) deviates from a straight line, which indicates that there is an additional molecular flow. The Knudsen number is $0.01 < Kn < 0.1$ for pressures between 1.3×10^3 and 0.1×10^3 Pa, which corresponds to the intermediate mode of flow for the mixture in a capillary, where the slip effect begins to make itself felt.

The pressure difference ΔP along a capillary [2] at atmospheric pressure ($v \sim 1 \cdot 10^{-9}$ m/sec for water) for a length $\Delta h = 0.1$ m ($r = 0.3 \cdot 10^{-3}$ m) was not more than 10^{-4} Pa, i.e., the pressure gradient is negligible, and the vapor leaves the meniscus only by diffusion and Stefan flow [3]. As the pressure is reduced, the capacity for those modes of flow is lowered [4] and the pressure difference increases. We estimated ΔP here on the basis of the slip and found that at 1.2×10^3 Pa, the difference for the same length was about 1×10^2 Pa, i.e., it was much larger than that at atmospheric pressure. The P_s here can then become higher than the gas pressure above the capillary, and we found for example that at 0.67×10^3 Pa and with the water at $T_\ell \approx 280$ K, $P_s \approx 10^3$ Pa, i.e., $P < P_s$.

This total pressure difference causes hydrodynamic motion, i.e., for $P < P_s$ the diffusion transport is replaced by molecular, which is described by Poiseuille's equation. In the intermediate range, where slip begins to occur, the rate of vapor loss from the capillary increases and the effects of menuscus depth on the evaporation rate are reduced (Fig. 4).

The cooling related to increased evaporation rate at low pressures should not affect the reduction in the rise because the surface tension increases as the temperature is reduced, while the wetting angle is reduced [2], i.e., the meniscus should rise, and should not show the observed descent.

The capillary and cell are communicating vessels with bends having different radii (capillary 0.30×10^{-3} m and cell 8.10×10^{-3} m). If there is no pressure difference in the capillary, the vapor leaves the meniscus or the liquid in the cell by diffusion, and the total pressure of the vapor and the air above the meniscus is equal to the pressure over the liquid in the cell, i.e., the environmental pressure P_e, so the meniscus height remains unchanged at pressures characterized by $Kn < 0.01$.

As the gas pressure is reduced, a difference in total pressure is set up along the capillary, which is due to evaporation from the meniscus and reduction in capillary carrying capacity, which alters the mechanism of vapor loss from the meniscus: diffusion is replaced by molecular transport. In the steady state, the total pressure at the meniscus exceeds that in the chamber, and thus is greater than that over the liquid in the cell ($P_e > P$), which causes the column in the capillary to descend, with a corresponding rise in the cell. The mean density within the capillary is $\rho_e = \rho_v + \rho_a$, which is greater than the density outside the capillary ($\rho_e > \rho$), and as the carrying capacity in viscosity-limited or effusive flow is dependent on the length and radius [4], the mean density is a function of those quantities, $\rho_e = f(L, r)$, so the meniscus step is dependent on the capillary length and radius.

The cell radius is about 30 times the capillary zone, and the evaporation will mean that the temperature in the cell is lower than that in the capillary, which is due to the recessed evaporation surface and the reduction in evaporation rate in the capillary consequent on it. Consequently, there is a difference in saturation vapor pressures over the meniscus and over the liquid in the cell because of the temperature difference, while the curvature in the capillary produces a vapor pressure difference opposite in sign, which to some extent compensates for that due to the temperature difference.

The meniscus oscillations accompanying the descent are due to the wetting angle varying drying a step. As the meniscus descends, it leaves behind it a film, so the wetting angle is reduced and the capillary pressure is increased, and thus the meniscus begins to rise again until the wetting film is restored and the contact angle reaches its equilibrium value, after which the above factors cause the meniscus to descend again. However, the rapid evaporation means that the thickness and length of the wetting film decrease over time [5], so the oscillations are damped and the meniscus halts at the position corresponding to the step in the given capillary.

Pressures less than those characterized by $Kn > 0.1$ do not alter the menuscus position, since in molecular flow the gas pressure difference in the capillary becomes quite small.

These measurements on capillary rise at reduced pressures make it necessary to consider the evaporation from the meniscus but show that there is a meniscus step, for which a physical explanation exists.

NOTATION

r and L, capillary radius and length; Δh, distance from meniscus to mouth of capillary; $\Delta\ell$, meniscus step; ℓ, meniscus rise; ℓ_∞ and x, maximal and relative rise; θ, wetting angle; P, gas pressure outside capillary; P_e, vapor—air mixture pressure above meniscus; ΔP, pressure difference in vapor—air mixture along capillary; P_s, saturation vapor pressure; T_ℓ, liquid temperature at meniscus; ρ gas density outside capillary; ρ_v, ρ_a, and ρ_e, densities of vapor, air, and vapor—air mixture; v, evaporation rate; Λ, molecular mean free path; Kn, Knudsen number.

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RELATIVE MOTION OF DROPS UNDER THE ACTION OF VARIABLE FORCES

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The boundary between "moderate" and "large" drops, which is fixed for each drop—medium system in gravitational conditions, shifts toward smaller drops with increase in the forces applied to the drop.

The relative motion of particles, including drops, in a medium is usually investigated in gravitational conditions, characterized by constancy of the forces applied to the particle over both time and space. The results of such investigations also from the basis for calculations in those cases where, according to the operating conditions of the apparatus, the forces acting on the particle differ from gravitational forces. The so-called standard drag curve is widely used; it consists of a dependence of the drag coefficient on the Reynolds variation over a wide range of variation of the latter. This curve is suitable for solid spheres, regardless of the nature of the applied forces and the physical properties of the medium. The situation is different for particles with a mobile interface with the medium. For bubbles and drops, the existence of a Reynolds number Re_b at which the drag coefficient begins to increase significantly with increase in Re has been established. The number Re_b, called the boundary or transition value, is assumed to be constant for each

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