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EXPERIMENTAL THERMOMOLECULAR PRESSURE RATIO OF HELIUM-3 DOWN TO 0.3°K

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A THERMOMOLECULAR pressure theory has been developed in detail by Weber¹ and has been used till now to calculate the corrections for the measurements of helium-3 and helium-4 vapour pressure. This theory was confirmed experimentally by several workers.²⁻⁶ In particular, Roberts and Sydoriak⁶ measured the ratio p_c/p_w for helium-3 down to 1.9°K (p_c and T_c are the pressure and temperature of the cold point, p_w and T_w those of the warm point). Only recently the problem of the thermomolecular pressure was raised again—by the

results of Edmonds and Hobson⁷ concerning the limiting value $p_c/p_w = (T_c/T_w)^{1/2}$, and by Watkins, Taylor, and Haubach⁸ in the intermediate and viscous regions for helium-3 and helium-4. The latter work is particularly interesting for us because the experimental data contained in it are comparable with those presented here. Papers of reference 7 and 8 and our present results show that Weber's theory is not satisfactory for an accurate calculation of the effect.

Before examining our results we will recall briefly a few elements of the theory. The most general equation obtained by the Weber's theory is the following:

$$\frac{dp}{dT} = \frac{K_1 6\eta^2}{\rho T} \cdot \frac{1}{R^2 + 4K_2 R \gamma_M} \dots (1)$$

where p is the pressure, T the temperature, R the radius of the pipe, η and ρ are the viscosity and the gas density, K_1 and K_2 are constants, and γ_M the slip coefficient of Maxwell's theory, which is of the same order of magnitude as the mean free path. By integrating equation (1), using expressions for η and ρ from the kinetic theory, one obtains the well known expression which states that the ratio of the extreme pressure p_c/p_w is only a function of T_c , T_w , R/λ_c , and R/λ_w (λ is the mean free path).

For helium-4 Weber and Schmidt have obtained the following relation by fitting the constants with experimental data

$$\log \frac{p_c}{p_w} = \frac{1}{2} \log \frac{T_c}{T_w} + 0.18131 \log \frac{Y_c + 0.1878}{Y_w + 0.1878} + 0.41284 \log \frac{Y_c + 1.8311}{Y_w + 1.8311} + 0.15823 \log \frac{Y_c + 4.9930}{Y_w + 4.9930} \dots (2)$$

where

$$Y = \frac{R}{\lambda} = \frac{R_p}{13.42} \left(\frac{273.15^\circ\text{K}}{T} \right)^{1.147}$$

TABLE 1

$T_c, ^\circ\text{K}$ ($\Delta T_c = \pm 0.005$)	$P_c, \text{mm.Hg}$	$P_w, \text{mm.Hg}$	P_c/P_w
0.805	3.03 ± 0.10	3.05	0.99
0.679	1.16 ± 0.05	1.35	0.88
0.645	0.85 ± 0.04	9.80 × 10 ⁻¹	0.88
0.590	0.49 ± 0.02	6.80 × 10 ⁻¹	0.74
0.511	0.19 ± 0.01	3.50 × 10 ⁻¹	0.55
0.467	(0.95 ± 0.09)10 ⁻¹	2.20 × 10 ⁻¹	0.43
0.440	(0.61 ± 0.06)10 ⁻¹	1.75 × 10 ⁻¹	0.34
0.370	(0.14 ± 0.02)10 ⁻¹	(8.58 ± 0.20)10 ⁻²	0.17
0.347	(0.80 ± 0.10)10 ⁻²	(6.40 ± 0.20)10 ⁻²	0.13

$R = 3.5 \times 10^{-2}$ cm, $T_w = 294 \pm 2^\circ\text{K}$

TABLE 2

$T_c, ^\circ\text{K}$ ($\Delta T_c = \pm 0.005$)	$P_c, \text{mm.Hg}$	$P_w, \text{mm.Hg}$	P_c/P_w
0.755	2.12 ± 0.08	2.14	0.99
0.695	1.33 ± 0.05	1.39	0.97
0.580	0.44 ± 0.03	5.20 × 10 ⁻¹	0.87
0.540	0.27 ± 0.02	3.70 × 10 ⁻¹	0.75
0.521	0.21 ± 0.01	3.00 × 10 ⁻¹	0.74
0.504	0.17 ± 0.01	2.55 × 10 ⁻¹	0.68
0.450	(0.73 ± 0.07)10 ⁻¹	1.35 × 10 ⁻¹	0.55
0.416	(0.38 ± 0.04)10 ⁻¹	(9.20 ± 0.10)10 ⁻²	0.41
0.397	(0.27 ± 0.03)10 ⁻¹	(6.80 ± 0.10)10 ⁻²	0.39
0.378	(0.17 ± 0.02)10 ⁻¹	(5.30 ± 0.10)10 ⁻²	0.32
0.370	(0.14 ± 0.02)10 ⁻¹	(4.40 ± 0.10)10 ⁻²	0.32
0.360	(0.11 ± 0.02)10 ⁻¹	(4.05 ± 0.15)10 ⁻²	0.27
0.349	(0.84 ± 0.13)10 ⁻²	(3.30 ± 0.05)10 ⁻²	0.26

$R = 9.0 \times 10^{-2}$ cm, $T_w = 294 \pm 2^\circ\text{K}$

TABLE 3

$T_c, ^\circ\text{K}$ ($\Delta T_c = \pm 0.005$)	$P_c, \text{mm.Hg}$	$P_w, \text{mm.Hg}$	P_c/P_w
0.755	2.12 ± 0.08	2.14	0.99
0.695	1.33 ± 0.05	1.36	0.98
0.580	0.44 ± 0.03	5.10×10^{-1}	0.89
0.529	0.23 ± 0.02	2.90×10^{-1}	0.85
0.521	0.21 ± 0.01	2.70×10^{-1}	0.83
0.506	0.17 ± 0.01	2.30×10^{-1}	0.77
0.450	$(0.73 \pm 0.07)10^{-1}$	$(1.20 \pm 0.01)10^{-1}$	0.62
0.416	$(0.38 \pm 0.04)10^{-1}$	$(7.00 \pm 0.04)10^{-2}$	0.54
0.397	$(0.27 \pm 0.03)10^{-1}$	$(5.60 \pm 0.20)10^{-2}$	0.48
0.375	$(0.16 \pm 0.02)10^{-1}$	$(4.10 \pm 0.10)10^{-2}$	0.40
0.370	$(0.14 \pm 0.02)10^{-1}$	$(3.60 \pm 0.10)10^{-2}$	0.39
0.359	$(0.11 \pm 0.02)10^{-1}$	$(3.00 \pm 0.10)10^{-2}$	0.36
0.348	$(0.82 \pm 0.13)10^{-2}$	$(2.65 \pm 0.10)10^{-2}$	0.33

$$R = 1.55 \times 10^{-1} \text{ cm}, T_w = 294 \pm 2^\circ\text{K}$$

TABLE 4

$T_c, ^\circ\text{K}$ ($\Delta T_c = \pm 0.005$)	$P_c, \text{mm.Hg}$	$P_w, \text{mm.Hg}$	P_c/P_w
0.759	2.18 ± 0.08	2.20	0.99
0.673	1.10 ± 0.05	1.13	0.97
0.645	0.85 ± 0.04	8.70×10^{-1}	0.97
0.511	0.19 ± 0.01	2.25×10^{-1}	0.85
0.482	0.12 ± 0.01	1.65×10^{-1}	0.78
0.451	$(0.74 \pm 0.07)10^{-1}$	1.07×10^{-1}	0.71
0.427	$(0.48 \pm 0.04)10^{-1}$	$(8.00 \pm 0.20)10^{-2}$	0.62
0.383	$(0.20 \pm 0.02)10^{-1}$	$(4.10 \pm 0.03)10^{-2}$	0.49
0.347	$(0.80 \pm 0.10)10^{-2}$	$(2.20 \pm 0.02)10^{-2}$	0.37
0.333	$(0.54 \pm 0.08)10^{-2}$	$(1.73 \pm 0.02)10^{-2}$	0.32
0.321	$(0.38 \pm 0.06)10^{-2}$	$(1.50 \pm 0.05)10^{-2}$	0.26
0.317	$(0.34 \pm 0.06)10^{-2}$	$(1.30 \pm 0.05)10^{-2}$	0.26

$$R = 2.4 \times 10^{-1} \text{ cm}, T_w = 294 \pm 2^\circ\text{K}$$

(R in cm and p in μHg). This is the equation used by Roberts and Sydoriak in their paper. With the help of an electronic computer, they have determined the ratio p_c/p_w for different T_c and T_w and for different values of Rp . These evaluations, as said before, have been confirmed by their measurements in helium-3 down to 1.9°K , while they are not in good agreement with experimental data obtained by Watkins et al.⁸ and those described in the present work.

The measurements reported here have been the subject of a doctoral thesis by one of us (A.F.) discussed at the University of Rome in 1965. A preliminary report appeared as an internal report.⁹

Experimental technique and results

To measure the thermomolecular pressure, we preferred an experimental situation in which p_c is the vapour pressure of a liquid helium-3 bath. In this way the measure of p_c becomes a measure of T . However, we will show that the knowledge of p_c is in fact independent from that of T_c . In this case it is not necessary to make any correction for adsorption of helium-3 on the walls since the bath acts as an infinite gas source. On the other hand, in order to have more data for the same T_c (and then the same p_c) it is necessary to change the radius of the pipe. We used stainless steel pipes of 5 different diameters: $2R = 0.48, 0.31, 0.18, 0.07, 0.03$ cm, 95 cm long. The measurements with the last pipe are not very reliable for obvious experimental difficulties: to have meaningful data equilibrium times needed to be about 2 h at the lowest temperatures; the data obtained are in substantial qualitative agreement with the whole set of measurements, but will not be reported here.

The diameters chosen cover, for every temperature, a range of Rp_w large enough to fit our data in Weber's theory through the calculations made by Roberts and Sydoriak.

The apparatus consists of a copper cell in which the helium-3 is condensed† containing two carbon resistor thermometers immersed in the liquid, one used for thermoregulation and the other for the measurement of temperature. The cell is soft soldered to a helium-3 refrigerator and communicates with the room temperature section through three pipes of different diameters in parallel: the measurement of p_w is made alternatively for each of the three pipes with the same manometers, in the same conditions of temperature and quantity of condensed helium-3. The measurements were taken in different runs, combining the five diameters chosen, three at a time, so as to check the data obtained.

T_c and p_c were measured by two Allen and Bradley carbon resistor thermometers (10 ohm, $\frac{1}{2}$ W) which were calibrated against the helium-3 vapour pressure measured statically in the helium-3 refrigerator. The helium-3 pumping line has a diameter of 12 mm and the correction for thermomolecular pressure is rather small, down to 0.4°K . Since the thermometers were immersed in a helium-3 bath differing from the bath whose vapour pressure was measured, a correction was made to take

† We have used helium-3 of the following purity: 99.86% total helium and 100.000% helium-3 in helium, supplied by Monsanto Research Company.

into account the thermal boundary resistance, following the data given by Fairbank and Lee.¹⁰ Once corrected the data fitted very well the relation

$$\log R = a - b \log p \quad \dots (3)$$

obtained by Cunsolo et al.¹¹ We assume, on the basis of the experimental evidence from Cunsolo et al., our own evidence, and the theoretical interpretation by Craig¹² that equation (3) is valid down to the temperatures that we reached. This hypothesis is very useful because it allows us to know p_c independently from the vapour pressure curve.

In order to evaluate with the maximum care the errors for p_c we compared the corresponding T_c with the results of Cunsolo et al.¹¹ (we want to point out that, for an evaluation of the errors, the absolute value of T_c is not significant). According to these authors and the dispersion of our own data, it seems reasonable to assume that the maximum error in the determination of the temperature was $T = \pm 0.005^\circ\text{K}$. From this, the error in the value of p_c can be determined. During the measurements the temperature T_c was kept constant with the help of an electronic thermoregulator driven by one of the two carbon resistor thermometers in the cell. The ΔT due to thermoregulation was certainly less than $5 \times 10^{-4}^\circ\text{K}$.

In Tables 1-4 we have collected the experimental data for the 4 tubes used. The errors in the measurement of p_w were shown by the calibration of the manometers (we

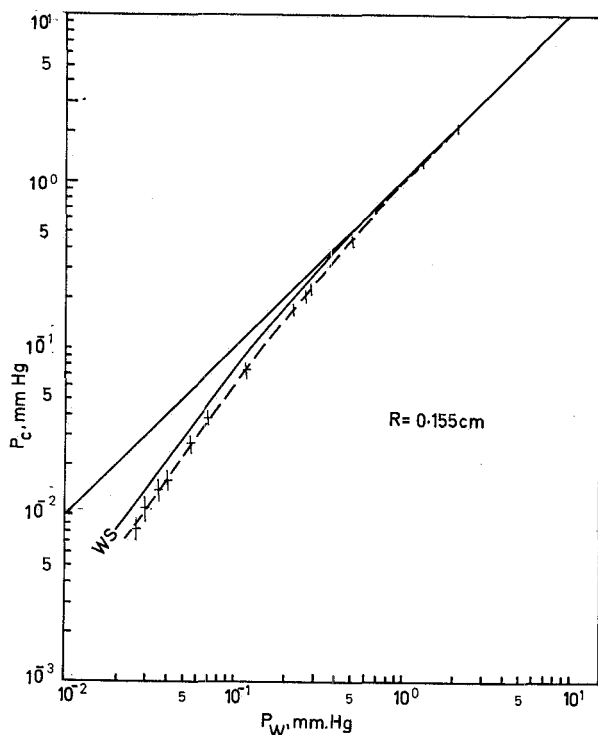


Figure 1. Helium-3 thermomolecular pressure for a tube of 0.155 cm radius. $T_w = 294 \pm 2^\circ\text{K}$; p_c is the vapour pressure of the bath and gives T_c directly (the T_c range is from ≈ 0.35 to 0.8°K). The straight line is $p_c = p_w$, i.e. absence of thermal pressure; the line WS represents equation (2); the points with errors and dotted line are the present measurements

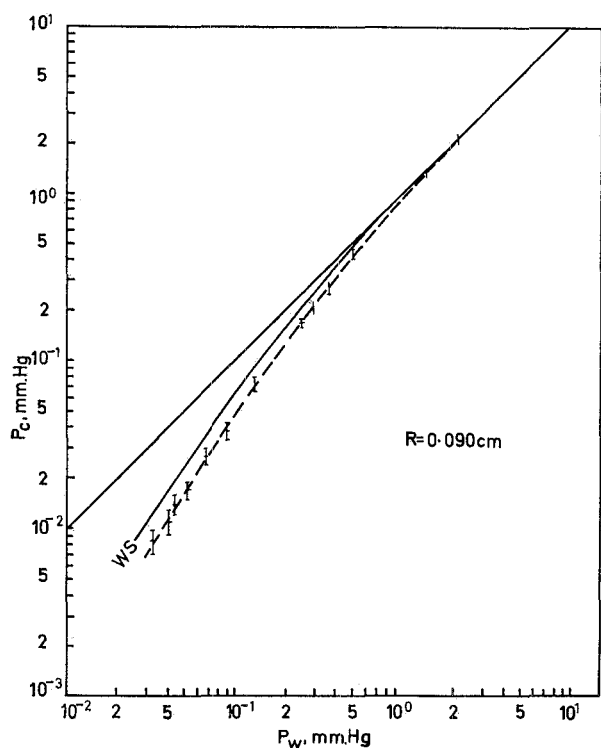


Figure 2. Helium-3 thermomolecular pressure for a tube of 0.090 cm radius; for the specifications see Figure 1

used a set of three manometers: an Hg, an oil manometer, and a McLeod with two capillaries of different sensitivities, all calibrated in a conventional way) and by the precision in reading (this was often improved by taking the average of repeated measurements); these fall within 0.5% for most of the data but where the error is larger than 0.5%, it is shown in the tables.

Discussion

Comparison with Weber's theory. In Figures 1-3 we have reported our experimental data in a plot of p_c versus p_w for different radii of the pipes: $R = 0.155$, 0.090 , and 0.035 cm. In the same graphs the theoretical curves calculated from Weber and Schmidt's equation (2) for pipes of the same radii are shown.

It is evident that the differences between calculated and experimental values are not favourable to the experimental errors, which were calculated in a pessimistic way. These differences cannot even be attributed to an error in the measurement of p_c , e.g. to a bad calibration of the thermometers; we found that, to fit our data on the calculated curves, we had to correct T_c in a way which was different for each diameter of the pipes.

We have tried to justify these differences in the scheme of Weber's theory, taking into account the non-ideality of helium-3 at the lower temperatures. We calculated p_c/p_w again using equation (2) down to a given temperature; below this temperature we integrated directly equation (1) using for η and ρ the data existing in the literature. Both η and B (the second virial coefficient, used to calculate ρ) have been computed from the work of Keller et al.¹³⁻¹⁵ and are in good agreement with the measurements of Becker et al.¹⁶ These authors, in order to obtain the values of η and B , used two different kinds of potential: the Lennard-Jones 12-6 (which they call LJ1) and an exp-6 potential (indicated by MR5). We used both sets of values, with a numerical integration of equation (1) which started from T_c rising to 2.2°K ; from this point on we used equation (2) up to $T_w = 294^\circ\text{K}$. The values so obtained are listed in Table 5 in the columns $p_w(\text{LJ1})$ and

TABLE 5. P_w VALUES CALCULATED FOR T_c AND p_c (GIVEN IN THE FIRST AND SECOND COLUMNS) FROM EQUATION (2) AND EQUATION (1) IN DIFFERENT WAYS (SEE IN THE TEXT) AND CORRESPONDING $P_w(\text{EXP.})$

$T_c, ^\circ\text{K}$	$p_c, \mu\text{Hg}$	$p_w(\text{RS})$	$p_w(\text{MR5})$	$p_w(\text{LJ1})$	$p_w(\text{LJ1})^*$	$p_w(\text{exp.})$
0.35	8.62	37.51	38.45	39.09	47.51	65
0.36	11.17	43.98	—	—	52.46	73
0.37	14.30	51.25	—	—	58.36	82
0.38	18.11	59.43	—	—	64.98	91
0.40	28.12	78.64	78.97	79.24	82.66	113
0.45	72.69	147.53	147.76	147.85	149.10	190
0.55	308.54	421.41	421.90	421.90	421.60	470
0.60	544.49	663.72	664.69	664.69	663.74	700
0.65	893.09	1 009.36	1 009.76	1 009.76	1 009.03	1 030

* $R = 3.5 \times 10^{-2}$ cm, $T_w = 294 \pm 2^\circ\text{K}$.

$p_w(\text{MR5})$. Furthermore, we used the same values to integrate equation (1) up to 10°K , still using equation (2) from this point up to room temperature; these values are also listed in Table 5, in the column $p_w(\text{LJ1})$.

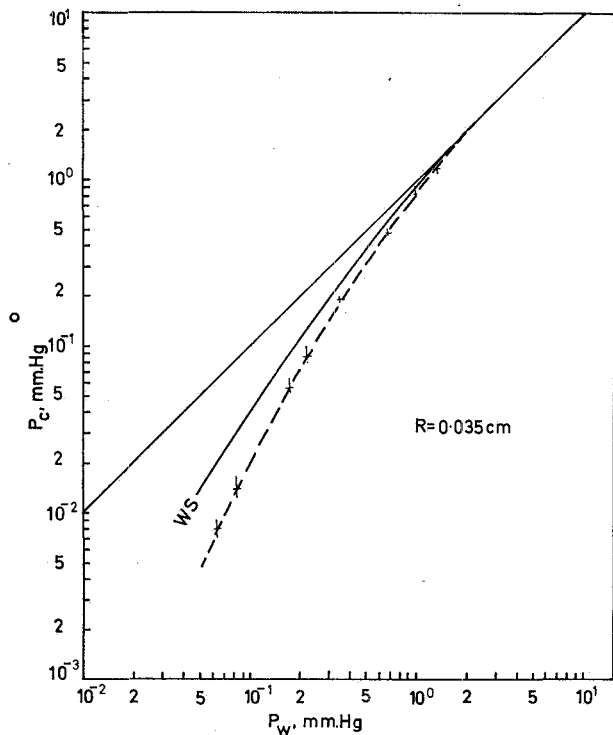


Figure 3. Helium-3 thermomolecular pressure for a tube of 0.035 cm radius; for the specifications see Figure 1

From Table 5 it is clear that taking into account the real behaviour of helium-3 in equation (1) did not eliminate the sensible differences between the calculated and the experimental values; it must be pointed out however that the correction acted in the proper direction, since the corrected values come closer than the uncorrected ones to the experimental data.

At this point one is tempted to make a deeper criticism of the Weber's theory. This is also justified by the following observation about our data. As said before, referring to equation (2), the ratio p_c/p_w is a function of the variables Rp_c , Rp_w , T_w , and T_c . Looking at our experimental procedure, if we keep T_c and T_w constant, p_c is also fixed. In this case p_c/p_w is a monotonous single-valued function of Rp_w . If we choose any value of the ratio p_c/p_w it follows that $Rp_w = \text{constant}$. In Figure 4 we have plotted the hyperbole obtained from equation (2), as calculated by Roberts and Sydoriak (in fact, the data are taken at slightly different temperatures in the range of T_c of 0.34–0.41°K), for the 4 radii which we used (full line). We have also deduced from our data the values of R and p_w which we obtain by keeping $(p_c/p_w)_{\text{exp}}$ fixed in the same temperature interval for T_c and taking one normalization point with the above curve (dotted line). It is easily seen that we do not obtain $(Rp_w)_{\text{exp}} = \text{constant}$. The data fit very well a straight line in a log R versus log p_w plot and the empirical relation that is obtained is $R^n p_w = \text{constant}$, with $n = 1.25$. This result, we hope confirmed by other measurements, demonstrates that it is no longer possible to use Weber's theory by a better fitting of the constants of equations (2), but that the theory must be modified in its analytical structure.

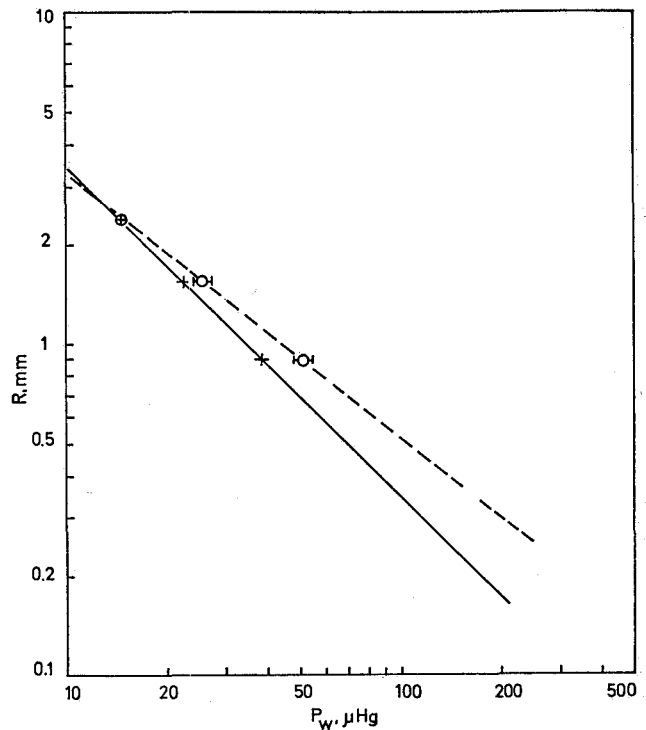


Figure 4. $R-p_w$ relations for $p_c/p_w = \text{constant}$ from equation (2) (full line) and from present experiment (dotted line)

Comparison with other experimental results. We note that it is not possible to overlap our data with those obtained by Roberts and Sydoriak.⁶ Measuring the vapour pressure of a liquid bath, we dealt with much higher pressures than those met by Roberts and Sydoriak; this means that at temperatures $T_c \approx 1^\circ\text{K}$ we find that the ratio $p_c/p_w \approx 1$, i.e. a thermomolecular effect is not measurable.

On the contrary we can compare our data with those obtained by Watkins et al.⁸ They are obtained measuring the difference between the pressures $p_w(r)$ of two pipes of different radius, which end in the same cell, with the same T_c and p_c . Since it is possible to write $p_w(r) = p_c + \Delta p(r)$, the measured quantity is $p_w(r_1) - p_w(r_2) = \Delta p(r_1) - \Delta p(r_2) = \Delta\Delta p(r_1, r_2)$. In this way the authors measure $\Delta\Delta p(r_1, r_2)$ as a function of $p_w(r_1)$, T_w , and T_c for several pipes and keeping T_c in the regions of liquid helium and nitrogen.

The results of these measurements are surprising because they do not agree with the values calculated from equation (2) not only at liquid helium but also at liquid nitrogen temperatures for the cold end. The measured thermomolecular effect is larger than the theoretical one and agrees with our results for T_c in the liquid helium range. We have interpolated our data (in the way shown in Figure 6) in order to obtain p_c versus p_w plots for the radii used by Watkins et al.; we have then calculated $\Delta\Delta p(r_1, r_2)$ for $r_1 = 0.29$ and $r_2 = 0.14$ which gives a value of 0.055. The agreement is good within the experimental errors.

The agreement between measurements obtained with different experimental procedures seems to indicate the opportunity to revise Weber's theory.

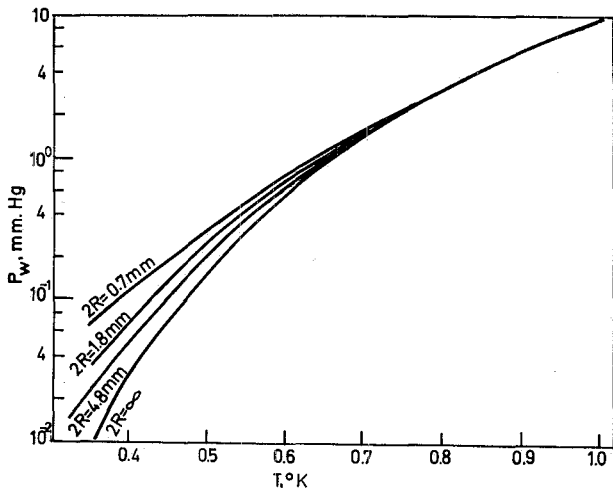


Figure 5. p_w - T_c curves from experimental points, for three pipe radii and $T_w = 294 \pm 2^\circ\text{K}$. The curve $R = \infty$ is the vapour pressure curve of Roberts and Sydoriak

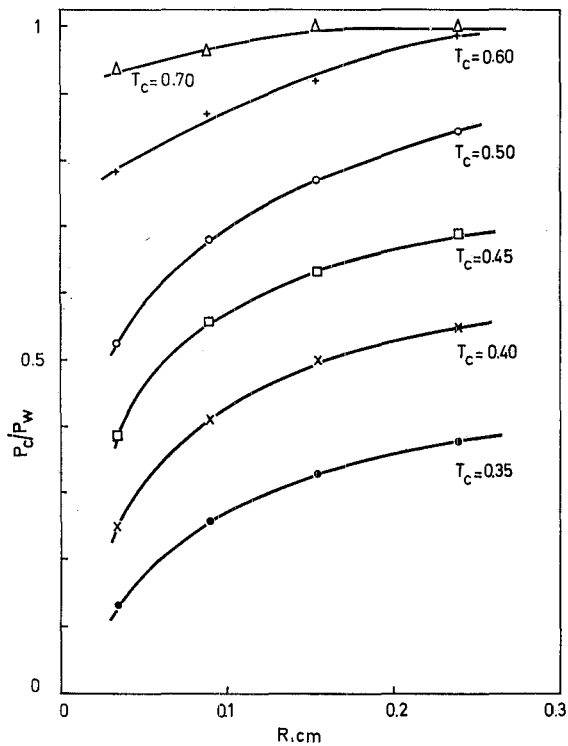


Figure 6. p_c/p_w ratios versus pipe radius R for several cold temperatures and fixed $T_w = 294^\circ\text{K}$

Use of the present data

With our experimental data we have drawn the graphs of Figures 5, 6, and 7, which can be used immediately to make thermomolecular pressure corrections as function of T_c and R . Figure 5 allows the direct reading of the temperature if one uses a liquid helium-3 bath connected to the room temperature manometer through a pipe whose radius equals one of those used by us. Figure 6 allows interpolations to make corrections for pipes of different diameters. We would not advise making extrapolations out of the range of diameters which we

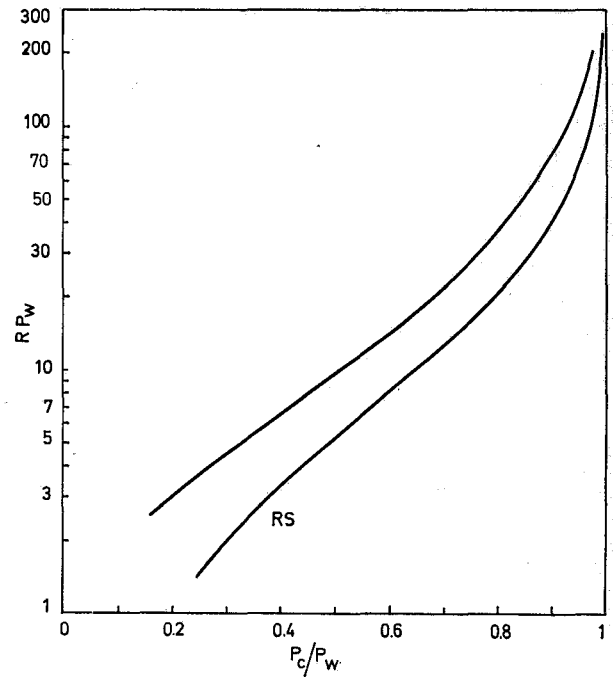


Figure 7. p_c/p_w ratios versus Rp_w (cm microns of Hg) product. RS curve is drawn for $T_w = 293.15^\circ\text{K}$ and $T_c = 0.4^\circ\text{K}$ from equation (2) using Roberts and Sydoriak⁶ numerical computation. The other curve is the best fit of our measurements between $T_w = 294 \pm 2^\circ\text{K}$ and $0.32 < T_c < 0.55^\circ\text{K}$

explored. Figure 7 shows another way to compare values calculated from equation (2) and our data. The ratio p_c/p_w is plotted as a function of the product Rp_w , for the same temperature difference. More exactly the theoretical curve (RS) is for $T_w = 293.15^\circ\text{K}$, $T_c = 0.4^\circ\text{K}$; our data are taken with $T_w = 294 \pm 2^\circ\text{K}$, $T_c = 0.55 \div 0.32^\circ\text{K}$ (it is the best fit computed for all our measurements contained in the quoted T_c interval). Although, as we have remarked in referring to Figure 4, this kind of plot is not in our opinion correct, we have preferred to report it in order to allow a rough correction, mainly for radii out of the range defined by the pipes that we used, for which Figure 6 cannot be used. We believe that in the curves drawn in these two figures the errors should be of the order of 8–10%. In Figure 5 the curve of $R = \infty$ is obviously the vapour pressure curve taken from the Roberts and Sydoriak T_{62} temperature scale.¹⁷

Conclusions

Two problems emerge from the measurements of Watkins et al. and our own. The first one is concerned with the opportunity of revising the thermomolecular pressure theory; this, in our opinion, should require a deeper analysis of the process of gas-surface interaction. For this it is necessary to have a better knowledge of the parameters involved, such as the effective cross-section, the accommodation and slip coefficients.

The second problem concerns the correction of the helium-3 vapour pressure curve. It should concern only the lower temperature region where the correction for thermomolecular pressure made with our data is different from that obtained with Weber-Schmidt equation (2).

Nevertheless, we think that it would not be wise to perform a correction with the data reported here since, as mentioned in discussion of equation (3), we lack a determination of T_c independent from vapour pressure (such as that measured with a paramagnetic salt). We want to emphasize that our data give only a relation between p_c and p_w .

The authors want to thank Dr. Buonanni and Dr. G. Verri for the numerical computations.

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CORRIGENDA

Vol. 7, No. 5. 'Transport Properties of Cryogenic Liquids and Their Mixtures.' G. T. Preston, T. W. Chapman, and J. M. Prausnitz.

Pages 275, 276: Figures 1-3 show data for carbon monoxide not carbon dioxide.

Vol. 7, No. 6. 'Flux Linkage and Self-Inductance of Superconducting Solenoids.' V. V. Sytchev, V. B. Zenkevitch, V. V. Andrianov, and F. F. Ternovskiy.

Page 345: In Table 1 figures for 'Critical current, A' should read

$$> 35\ddagger \quad 22.0 \quad > 35\ddagger$$

Page 346: Column 1, line 8 should read internally linked flux. The current dependence of ψ_i can

Page 347: Column 1, line 23 should read which magnetic induction in the centre of the superconducting

Column 2, line 3 should read

wire material and increases with an increase of structural

Column 2, line 8 should read

characteristic of the wire of solenoid 3. For a finite solenoid H^* is an

Page 348: Column 2, lines 49, 50 delete which is characteristic of the discharge at infinitely high current

Page 349: Caption to Figure 7 should read Current dependence of static inductance L^{st} of solenoid 1

Equation (12) should read

$$L^{st} = \frac{\psi - \psi_0^{\max}}{I}$$

Column 2, line 15 should read

consideration does not undergo any substantial change

Column 2, line 18 should read

inductance differs from the nonmagnetic solenoid inductance

Page 350: Reference 1 should read

SYTCHEV, V. V., ZENKEVITCH, V. B., and ANDRIANOV, V. V. *Doklady AN SSSR* **165**, 73 (1965)

Reference 2 should read

ANDRIANOV, V. V., ZENKEVITCH, V. B., SOKOLOV, V. I., SYTCHEV, V. V., TOUMA, V. A., and FEDOTOV, L. N. *Doklady AN SSSR* **169**, 316 (1966)

Vol. 7, No. 6. 'A Chamber for Working at Liquid Helium Temperatures up to 18 kbar.'

Authors should read

E. S. Itskevich, A. N. Voronovskii, A. F. Gavrilov, and V. A. Sukhoparov.