CONVECTION AND HEAT TRANSFER NEAR THE CRITICAL POINT OF CARBON DIOXIDE

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A study has been made of convective heat transfer between carbon dioxide and electrically heated platinum wires in horizontal and vertical channels. The heat-transfer and convection coefficients have been found and a dimensionless correlation determining the onset of convection verified.

It was observed long ago that, in the presence of gravity, when a fluid is near the critical state, a small temperature inhomogeneity is sufficient to cause convective currents. This is due to the high value of the thermal expansion coefficient near the critical point.

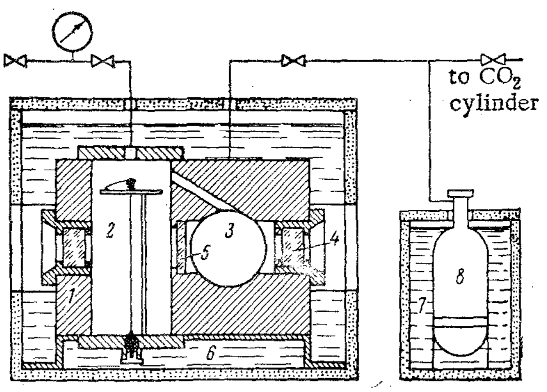

Fig. 1.

The intensification of natural convection in a homogeneous system in the near neighborhood of the critical point is of considerable interest in two respects. First, this effect intensifies heat transfer inside the fluid and from the solid wall [1, 2] in the case of small temperature differences. Second, in determining the true coefficient of molecular thermal conductivity λ one must allow for the enhanced possibility of the onset of natural convection.

It is well known that most thermophysical and kinetic properties of a fluid (specific heat capacity, compressibility, thermal expansion, absorption, speed of sound, and diffusion coefficient) pass through extrema in this region. Of these, thermal conductivity is the most difficult to measure due to the influence of convection. The question of the existence of maxima of thermal conductivity has been debated for more than 20 years [3-6]. Theoretical arguments supporting the existence of an extremum of λ in the near supercritical region are given in [7].

We have studied the relation between the conditions that facilitate the onset of convection and the nature of heat transfer for a wide range of states. Particular attention has been paid to states on both sides of the line of supercritical continuous transitions [8] through a region of reduced thermodynamic stability of the phase.

In addition to determining the heat transfer coefficient α, we calculate the effective thermal conductivity λ^* and the ratio $\lambda^*/\lambda = \varepsilon$. If at some pressure the value of ε on the supercritical isotherm passes through a maximum, then the convective component of the transport coefficient must grow faster than the molecular component, even though the latter also passes through a maximum. The convection pattern near heated vertical and horizontal wires in channels was studied visually and photographed by means of an "Admira-8PA" motion-picture camera or a "Start" still camera.

The experimental setup is shown schematically in Fig. 1. Chamber 1 is made of stainless steel. The vertical and horizontal channels 2 and 3, which contain the heating elements, have identical dimensions (channel length 120 mm, diameter 40 mm). Visual observations and photographs were made through self-sealing glass windows 4, designed to withstand pressures of more than 100 kg/cm^2, and an unloaded glass window 5 which separated the channels. A 29μ diam. platinum wire, mounted on the flange, served as a heating element and resistance thermometer. The length of each wire was about 80 mm, and its surface area was F = 7.2-7.4 mm^2 (the wires had to be replaced several times).

The chamber was totally immersed in a 15-liter water thermostat 6. The prescribed temperature was maintained in the thermostat to within $\pm$0.01°C. During most of the experiments the state of the fluid changed along an isotherm. A second thermostat 7 held a 1.3-liter steel bottle 8 containing carbon dioxide at sufficiently high density. This thermostat was used for thermal regulation of the pressure. The system could be easily divided into parts by means of four needle valves. For example, after the desired pressure was attained, the chamber could be separated from the rest of the system. Pressure was measured by means of a class 0.35 spring manometer with a 250 kg/cm^2 full scale. The carbon dioxide used contained about 0.3 % inert impurities. Current for the electrical circuit was supplied from a bank of acid batteries. The rate of heat generation and wire resistance r were calculated from the voltage drops across the wire and across a reference resistance.

The thermal coefficient of resistance B was determined for each of the two wires in the temperature range 30-40°C by means of a mercury-in-glass thermometer with 0.1 °C scale division. The temperature of the experiment exceeded these limits only in a few cases (e.g., in helium experiments). In these cases the coefficient B was measured in the appropriate range.

Before the heat transfer coefficients were measured, the chamber was brought to the prescribed temperature. The value of the wire resistance r_0, which corresponded to the temperature of the experiment, was found by graphical extrapolation of the $r = f(i^2)$ relation in r, i^2 coordinates to zero current. Either the vertical or the horizontal wire was projected on the screen. The field of vision in the chamber was a circle 15 mm in diameter. The light source was a 25W incandescent bulb. For photography the screen was replaced by a camera without an objective. Exposure time was 0.001 sec.

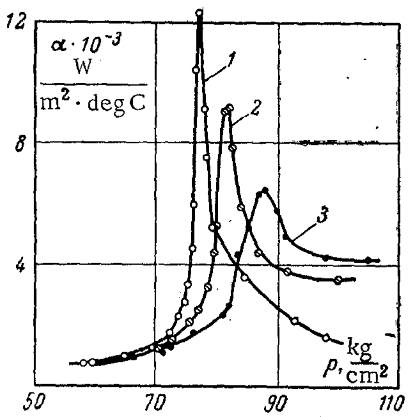

Fig. 2.

In the experiments we measured, in addition to temperature and pressure, the heat flux Q (or specific heat flux q) and temperature drop Δt, which were then used to calculate the heat transfer coefficient α. For small temperature drops $\Delta t = t_w - t_f$ (t_w – wire temperature, t_f – bulk temperature of fluid) the inhomogeneity of the fluid properties near the heater and in the bulk had practically no effect on the nature of the convective flow. The diameter D of the chamber channels was so large, as compared with the wire diameter d (D/d = =1380), that for the rate of heating used, 0.002-0.04 W over 80 mm length, the assumption of a bulk region in the fluid with temperature equal to the chamber-wall temperature was a reasonable idealization of the actual conditions. The main experiments with carbon dioxide in the supercritical region were performed with Δt = 0.5°C.

A series of isothermal curves $\alpha = \alpha(p)$ for the horizontal wire is shown in Fig. 2, where curves 1, 2, 3 correspond to t_1 = 31.5, 34.0, 37.0°C, respectively. The temperature of the experiments exceeded the critical temperature by 0.4° for curve 1, 3.0° for curve 2, and 6.0° for curve 3. The heat transfer coefficient α passes through a maximum, which decreases in magnitude and shifts toward higher pressures with increasing chamber temperature. The line joining the maxima of α in the p, t system of coordinates corresponds to an extension of the saturation curve beyond the critical point [2].

The heat transfer data for the vertical wire exhibit the same dependence on pressure and temperature as those for the horizontal wire, but the heat transfer coefficient is systematically lower.

The passage of the heat transfer coefficient through a maximum on supercritical isotherms is connected with the onset of turbulent convection near the heater and the existence of a sharp maximum of the specific heat capacity c_p at densities near the critical state. The influence of the various factors on the nature of the heat transfer is clearly expressed in the dimensionless equation

$$N = f(R) \qquad (N = \alpha l / \lambda) \qquad (1)$$

where R and N are the Rayleigh and Nusselt numbers, respectively.

TABLE 1

Specific heat flux q [W/m²] and heat transfer coefficient α [W/m²deg C] as a function of pressure along the 32.0°C isotherm for the horizontal and vertical wires.

p, kg/cm²	Horizontal			Vertical		
	Δt °C	q 10⁻³	α 10⁻³	Δt °C	q 10⁻³	α 10⁻³
61.1	0.50	0.41	0.82	0.49	0.30	0.625
66.7	—	—	—	0.50	0.43	0.86
67.8	0.48	0.50	1.05	—	—	—
70.3	0.47	0.57	1.21	0.51	0.55	1.08
73.5	0.48	0.80	1.66	0.51	0.67	1.32
75.5	0.48	1.26	2.63	0.50	0.96	1.92
76.8	0.49	1.90	3.87	0.51	1.68	3.29
78.0	0.47	4.06	8.64	0.53	2.64	4.98
78.3	0.50	5.90	11.80	0.50	4.40	8.80
79.0	0.47	4.05	8.62	0.51	2.82	5.53
82.0	—	—	—	0.48	1.58	3.29
83.7	0.51	1.73	3.38	—	—	—
103.0	0.47	1.28	2.73	0.53	0.90	1.72

Although in these experiments the channel diameter was three orders larger than the heater diameter, heat transfer from the wire cannot be regarded as convection in an infinite volume. The onset of convection is determined by the stability of a cylindrical column of fluid. The characteristic diameter of this column can be significantly larger than the wire diameter d. The use of the value d = 29 μ in the Rayleigh number leads to physically incorrect estimates: even for the regime of fully developed turbulent convection near the critical point we obtain overly low values of R, of the order of 10^3. If the channel diameter D = 40 mm is taken as the characteristic dimension, then the nature of the convective flow is in qualitative agreement with the order of the Rayleigh number. For example, in the case of the 32.0°C isotherm the heat transfer coefficient passes through a maximum at a pressure of 78.3 kg/cm². In this case R = $= 5.4 \times 10^{12}$ for Δt = 0.48°. Further away from the maximum the value of R on this curve is of the order of 10^8-10^9.

After this discussion we can pass to the representation of the experimental results by means of the convection coefficient ε. Inside the experimental cell the fluid is contained between two cylindrical surfaces: the outer surface of the wire and the inner surface of the channel. When heat is transferred only by molecular thermal conduction, the heat flux associated with a given temperature drop Δt can be calculated from the known formula.

Values of λ for carbon dioxide are given by Michels et al. [6]. On supercritical isotherms λ passes through a maximum.

In the presence of convection the heat flux Q is always greater than Q_λ. Using the effective coefficient of thermal conductivity, one can establish a functional relation between Q and Δt, but the similarity between λ and λ* is purely formal. The variable λ* refers essentially to a hypothetical fluid in which the experimentally observed heat flux Q

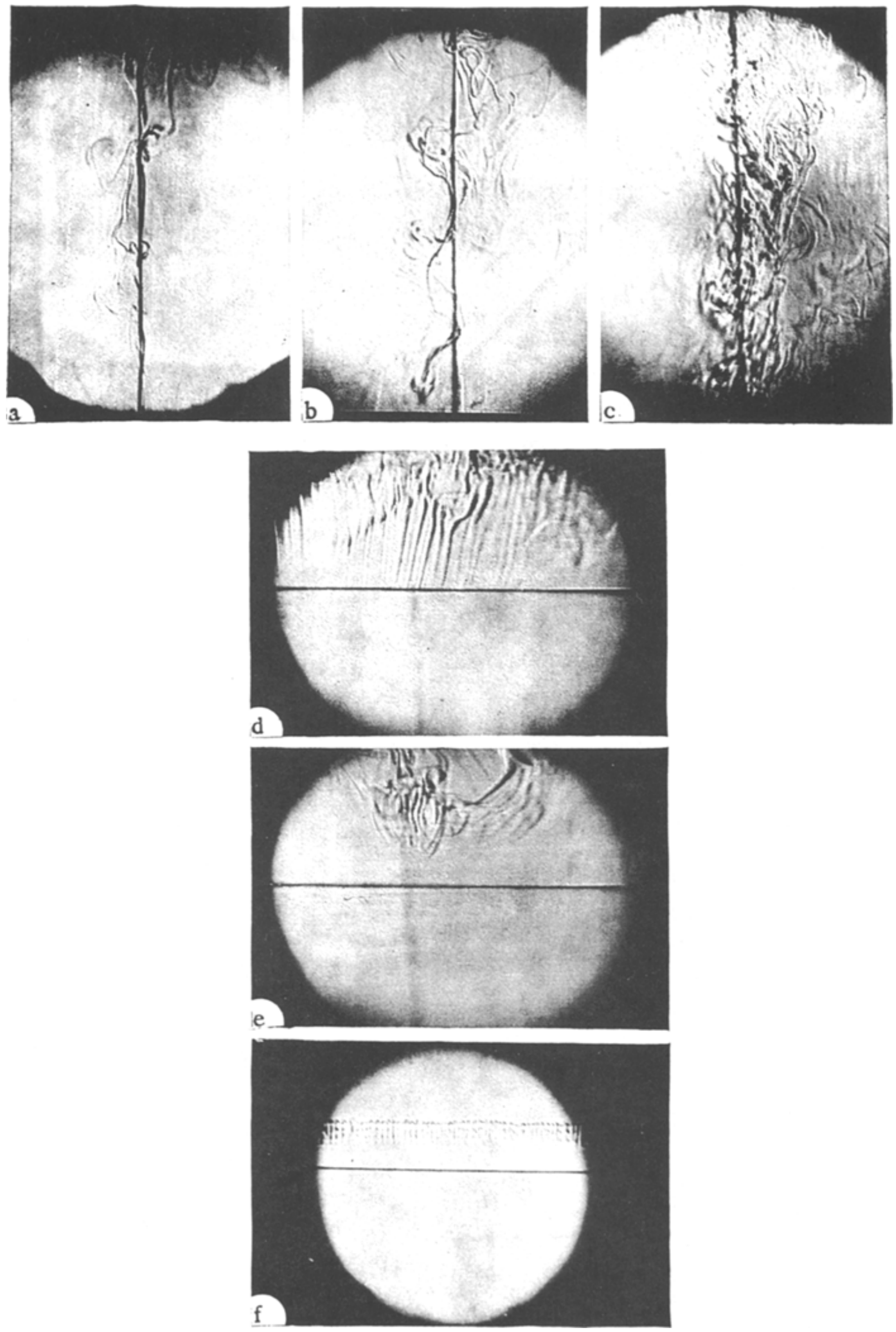

Fig. 3.

could be obtained with the given temperature drop Δt by pure thermal conduction. The convection coefficient is defined as

$$\varepsilon = \frac{\lambda^*}{\lambda} = \frac{Q}{Q_\lambda}.$$

Table 2 shows the measured values of Δt and Q and the calculated* values of Q_λ and ε for the horizontal channel with chamber temperature 32.0 °C and different pressures p (kg/cm²). It can be seen that the convection coefficient also passes through a pronounced maximum.

*The values of α and λ at the maximum points were correlated with respect to pressure.

The values of λ taken from [6] relate to the temperature 32.1°C. In our experiments the temperature outside the thermal boundary layer was 32.0°C, while at the surface of the heater it was 32.5°C, i.e., the mean temperature exceeded the reference value of [6] by 0.1-0.2°C. It has been shown that a decrease in Δt leads to a significant increase in λ^* only at the maximum point. Thus, at a pressure of 78.3 kg/cm^2 (Table 2) a change from $\Delta t = 0.5$°C to $\Delta t = 0.2$°C leads to $\varepsilon = 12.2$ instead of $\varepsilon = 9.2$. At some distance from the maximum the effect of Δt on λ^* becomes negligible, if the experiment is in the range of small Δt.

TABLE 2

Convection coefficient ε and heat fluxes Q and Q_λ [W] in carbon dioxide as a function of pressure p (kg/cm^2) along the 32.0°C isotherm. (Horizontal wire)

p	Δt °C	Q 10^3	Q_λ 10^3	ε
1.0	0.56	1.01	0.65	1.50
61.1	0.50	3.06	0.94	3.26
67.8	0.48	3.70	1.07	3.46
70.3	0.47	4.22	1.14	3.70
73.5	0.48	5.89	1.30	4.53
75.5	0.48	9.32	1.74	5.36
76.8	0.49	14.02	2.28	6.15
78.0	0.47	30.04	3.10	9.70
78.3	0.50	43.66	4.74	9.20
79.0	0.47	30.00	3.27	9.17
83.7	0.51	12.77	2.73	4.68
103.0	0.47	9.49	2.71	3.50

According to published estimates [9, 10], the condition for the absence of free convection is given by the inequality

$$R \leq 1000 \quad (2)$$

In our case we use the diameter of the inner channel D = 40 mm as the characteristic length in the Rayleigh number.

In the absence of convection $\varepsilon = 1$. Table 2 shows that in the case of carbon dioxide $\varepsilon > 1$ even at atmospheric pressure in spite of the small temperature drop. This agrees with the estimate, as in our case $R = 3.5 \times 10^4 \Delta t$ (t = 0°C, p = 1 kg/cm^2). To make the Rayleigh number less than 1000, we would have to keep Δt less than 0.03°C. Our setup did not permit such small temperature drops. As a check on the experimental procedure it was necessary to perform the experiment under conditions satisfying (2) and see whether the convection coefficient was in fact equal to unity.

A suitable gas for this experiment is helium at atmospheric or higher pressure. At t = 0°C, p = 1 kg/cm^2 the value of R is $1.40 \times 10^2 \Delta t$ and at t = 50°C, p = 1 kg/cm^2 we have $R = 1.45 \times 10^2 \Delta t$. Thus $R < 1000$ up to $\Delta t \approx 7$°C. The results of the experiments with helium are given in Table 3. In fact, we do have $\varepsilon = 1$.

If we regard the estimate (2) as valid up to the critical point, then in determining thermal conductivity by the method of coaxial cylinders we must use extremely small clearances b. For example, in the case of carbon dioxide at 32.0°C with $\Delta t = 0.1$°C and a pressure corresponding to the maximum of the heat transfer coefficient, the limit is $b \leq 0.1$ mm. With larger clearances, the results of measurements of the thermal conductivity near the maximum are affected by convection. Our experiments show that it is preferable to use vertical, rather than horizontal, cylindrical layers, since then, all other conditions being equal, the convective component of heat transfer is higher.

TABLE 3

Convection coefficient ε and heat fluxes Q and Q_λ [W] in helium for p = 1 kg/cm^2, t = 34.0°C as a function of the temperature drop Δt (Horizontal wire).

Δt °C	Q 10^3	Q_λ 10^3	ε
0.24	2.49	2.53	0.98
0.44	4.80	4.63	1.04
0.61	6.34	6.42	0.98
0.86	8.88	9.03	0.98
1.25	13.44	13.16	1.02
2.13	22.98	22.4	1.02
4.49	48.53	47.6	1.02
8.40	92.6	92.0	1.01
11.6	130.5	131.2	0.99

Let us discuss briefly the results of visual and photographic studies of the convection of carbon dioxide near the critical point. By adjusting the lenses we could focus part of the vertical or horizontal wire on the screen. With a temperature drop $\Delta t = 0.5$°C, no convective flow could be observed near the wire on the lower part of the supercritical isotherms (p = 62 kg/cm^2, Fig. 2). With increasing pressure we observed individual curved streams running along the vertical wire with a period of 10-12 sec. As the maximum of the heat transfer curve is approached, this period decreases and the flow takes on the form of eddies. Near the maximum the convective flow becomes distinctly turbulent with fine curls and a continuous flow near the wire.

With increasing Δt, the region of visible convection widens. Fig. 3 shows the convection patterns near the vertical and horizontal wires for t = 34.0°C, p = 81.7 kg/cm^2 and (a) $\Delta t = 1.2$°C, $q = 5.8 \times 10^3$ W/m^2, (b) $\Delta t = 2.7$°C, $q = 15 \times 10^3$ W/m^2, (c) = 13.4°C, $q = 52.3 \times 10^3$ W/m^2, and for t = 32.0°C, p = 78.3 kg/cm^2 and (d), (e) $\Delta t = 17.7$°C, $q = 72.7 \times 10^3$ W/m^2. Flow (f) was observed when the current was switched on at t = 34.0°C, p = 81.7 kg/cm^2, $\Delta t = 10.0$°C, $q = 40 \times 10^3$ W/m^2. After passing through the maximum, the convection pattern again has the form of eddies. Critical opalescence occurs on the 31.5°C isotherm.

The flow observed on the horizontal wire is quite different. Convective currents can be seen only near the maximum of the heat transfer coefficient, even in the case of the 31.5°C isotherm. In other cases the currents are invisible, even though $\alpha_2 > \alpha_b$. It is possible that the convective flow has the form of a film that loses stability and breaks down into separate streams outside the field of vision. Near the wire two alternating flow patterns could be observed: ordinary turbulent flow, and the more regular streamline flow visible in Fig. 3. Occasionally the flow completely disappears from the field of vision. Subsequently straight streamlines again rise at right angles to the wire, to be followed by typically turbulent convection with vortices.

An interesting pattern was observed on the horizontal wire at the moment the current was switched on. The flow separating from the wire has the form of a ribbon divided into small transverse cells (Fig. 3). This structure is due to the hydrodynamic instability of the hot moving "ribbon." In the case of a high temperature drop ($\Delta t = 56\,^\circ C$, $\alpha = 2500\ W/m^2 \cdot deg\ C$), cinematographic data show that the ribbon rises at a rate of 2 cm/sec.

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