# Heat Transfer to Non-Newtonian Fluids

# A. B. METZNER, R. D. VAUGHN and G. L. HOUGHTON

University of Delaware, Newark, Delaware

This paper presents the first theoretical analyses combined with an experimental study of the variables controlling heat transfer rates to non-Newtonian fluids in the streamline-flow region. The theoretical analyses, for the limiting types of non-Newtonian materials, were related to the intermediate case of Newtonian behavior to form a coherent theory applicable to Newtonian and non-Newtonian fluids alike.

The experimental data covered Graetz numbers between 100 and 2,000 and were correlated with a mean deviation of 13.5%. The flow-behavior indexes of the three non-Newtonian fluids used varied from 0.18 to 0.70.

Some preliminary non-Newtonian results are presented on the problems of nonisothermal fluid-flow pressure losses and heat transfer outside the laminar-flow region. Further theoretical work is recommended in both these areas. Additional experimental data would be of value in all of the problems discussed.

The industrial importance of non-Newtonian behavior is generally known and the types of non-Newtonian behavior encountered have been discussed by many authors (1 and 13, for example); nevertheless, not a single proved method is available for prediction of heat transfer rates to highly non-Newtonian fluids such as viscous slurries, gels, and polymeric melts and solutions. The object of this investigation was to develop a quantitative understanding of at least a part of this problem. The streamline-flow region was chosen for study to enable a rigorous theoretical analysis and to take advantage of the usual high consistency of these materials, which make the streamline region of primary importance in many industrial applications.

It has been shown (13) that previous experimental work in the area of heat transfer to suspensions and other non-Newtonian materials may be divided into two categories.

One category consists of publications devoted to a study of suspensions of relatively inert (i.e., nonsolvated) solids or of dilute suspensions of other solids. The work of Miller (16), Orr and Dalla-Valle (18), and Winding and coworkers (23) and parts of the work of Bonilla and coworkers (2) and of Salamone and Newman (21) fall into this category. Such suspensions are usually nearly Newtonian in behavior; hence the peculiarities due to the non-Newtonian properties are difficult to evaluate experimentally. Most of these investigators made some attempt to consider this problem, but the purposes of their work usually did not include the presentation of broad defining equations which might apply to highly non-Newtonian systems as well as to the inert suspensions studied. Accordingly, this part of the prior art sheds relatively little light on the problem under consideration here, although the publications are of considerable interest in the field for which they were primarily intended.

The second of the categories deals with empirical correlations of experimental data on more highly non-Newtonian systems. The publication of Chu and coworkers (5) and an appreciable part of the Bonilla et al. (2) and Salamone and Newman (21) data fall into this group. It has been shown elsewhere (12, 13) that the empirical correlations proposed by these authors are unrealistic and in two cases may even lead to prediction of negative heat transfer coefficients. Attempts to recorrelate the experimental results (4, 13) have been only partially successful, possibly owing to limitations of the rheological data. Accordingly, prior to this work, reliable heat transfer estimates could be made only if an industrial problem dealt with one of the few fluids which have been studied experimentally. This would enable use of the raw data rather than of any correlation, but such estimates would, of course, be limited to the specific experimental conditions which had been

As the over-all problem was not well understood, the need for as rigorous a theoretical approach as possible was indicated. Pigford (19) recently published the first theoretical study of heat transfer to non-Newtonians in the laminar-flow region. This work represents a generalization of Leveque's method (11) for predicting forced-convection heat transfer coefficients for Newtonian fluids.

Leveque's solution of the simplified Fourier-Poisson equation for heat conduction to a moving fluid assumes that the temperature-boundary layer for the important case of high mass flow rates through short tubes is confined to a thin region near the heated surface. In this region it may be assumed that the velocity varies linearly with the distance

from this surface. If this is true, the velocity distribution in this region may be represented by

$$u = \alpha(R - r) \tag{1}$$

where  $\alpha$  is the velocity gradient at the wall. Leveque's final result was

$$\frac{h_a D}{k} = 1.615 \left( \frac{\alpha C_P \rho D^3}{8kL} \right)^{1/3}$$
 (2)

For a Newtonian fluid  $\alpha=8V/D$  and Equation (2) reduces to the familiar Leveque solution where the term in brackets becomes the product of  $4/\pi$  and the Graetz number.

For a non-Newtonian fluid the value of  $\alpha$  differs from 8V/D but may be obtained from the Mooney-Rabinowitsch equation for flow in circular conduits (1, 14, 17, 20). Pigford has rewritten the Leveque equation in the form

$$\frac{h_a D}{k} = 1.75 \delta^{1/3} \left( \frac{w C_p}{k L} \right)^{1/3} \tag{3}$$

where

$$\delta = \frac{\alpha}{8V/D} \tag{4}$$

Physically the term  $\delta$  represents the ratio of the velocity gradients at the walls of the tubes and hence is related to the ratio of the heat transfer rates for a non-Newtonian fluid compared with that for a Newtonian. Pigford (19) showed that this ratio may be evaluated as follows, if the basic assumptions of the Leveque approach are valid, For Bingham plastic fluids:

$$\delta = \frac{1 - \tau_y / \tau_w}{1 - 4/3 \ \tau_y / \tau_w + 1/3 \ (\tau_y / \tau_w)^4}$$
 (5)

For pseudoplastic fluids:

$$\delta = \frac{3n' + 1}{4n'} \tag{6}$$

Evaluation of  $\delta$  by means of Equation (6) actually does not require assumption of pseudoplastic-fluid behavior. The term n' is the flow-behavior index of the fluid (13, 14) and quantitatively defines the behavior of any type of non-Newtonian material. Accordingly, Equation (6) may be used with Bingham plastic and dilatant fluids as well as with pseudoplastics.

As the limiting cases of "infinite," or ultimate, Bingham-plastic or pseudo-plastic behavior are approached,  $\tau_{\nu}/\tau_{w}$ 

R. D. Vaughn is at present with Shell Development Company, Emeryville, California.

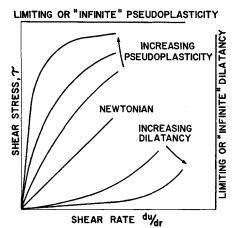


Fig. 1. Shear-stress-shear-rate diagram indicating various types of fluid behavior.

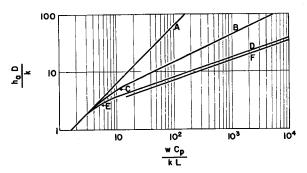
analytical solution may appreciably reduce the applicability of the results. A generally simpler and often as satisfying method of attacking such problems from an engineering point of view is to study the limiting cases which the problem may be shown to approach. This is the method of attack used in the present paper.

The limiting situations which encompass all types of steady state fluid behavior may be defined by reference to Figure 1. As a fluid becomes progressively more pesudoplastic, the shear stress-shear rate relationship progressively approaches the indicated horizontal line more closely. At this limit, the shear stress becomes independent of shear rate and the flow behavior index n' (13) approaches zero. At the other extreme of increasing dila-

pectic fluids would normally be reduced to conditions under which the time-dependent effects would no longer manifest themselves. Under such conditions design for these materials would be identical with the procedure outlined here for other types of non-Newtonian behavior.

The remainder of the theoretical development consists of two distinct steps: (1) development of theoretical equations for the cases of infinite pseudoplasticity (n' = 0) and infinite dilatancy  $(n' = \infty)$  and (2) development of a method of interpolation for cases of finite degrees of non-Newtonian behavior. This second problem is appreciably simplified by virtue of the extensive analyses available for the intermediate case of Newtonian behavior (n' = 1.00).

# Fig. 2. Summary of theoretical heat transfer relationships for various types of fluid behavior; curve CB: plug flow ("infinite" pseudoplasticity); curve ED: parabolic velocity profile (Newtonian fluid); curve F: conical velocity profile ("infinite" dilatancy).



approaches a value of unity and n'approaches zero. Accordingly, the values of  $\delta$  predicted by Equations (5) and (6) approach infinity, causing the heat transfer coefficients predicted by Equation (3) to do the same. The source of this difficulty lies in the assumptions underlying the Leveque approximation. In either of these limits, plug flow is approached (13, 22), and hence the region near the wall for which the velocity distribution may be assumed linear [Equation (1)] becomes smaller than the region in which there is a temperature gradient. Accordingly, the velocity gradient in the fluid near the wall has ceased to be a determining factor in the rate of heat transfer; hence the basic premise of the Leveque approach no longer is valid. In the limit of complete "plug" flow of the fluid the problem is identical to that of conduction into a solid cylinder initially at a uniform temperature. The rate of heat transfer in this case is determined solely by the rate of molecular conduction. As this can give only a finite rate of heat transfer, the foregoing equations rather evidently need some modifications near this limiting case of plug flow.

# THEORETICAL DEVELOPMENT

In many studies of complex problems the mathematical equations describing the physical situation are too difficult to solve, and the number of simplifying assumptions necessary to obtain an tancy, the vertical asymptote is approached as n' approaches infinity. Intermediate between these two situations of ultimate or "infinite" pseudoplasticity and infinite dilatancy lies the Newtonian fluid (n' = 1.00).

It may be noted in passing that the case of an "infinitely" Bingham-plastic fluid is indentical with that of infinite pseudoplasticity. As the method of approach used here for pseudoplastic fluids is also rigorously applicable to Bingham-plastic behavior and in view of the greater generality of the pseudoplastic-Newtonian-dilatant family of flow relationships, the case of heat transfer to true Bingham-plastic fluids, which are encountered only rarely, will not be presented in detail here. However, this case was treated analogously and an experimental verification of the theoretical relationships was made for one fluid approximating Bingham-plastic behavior. These results are available (22).

Thixotropic and rheopectic (13) non-Newtonian fluids, which have time-dependent properties, cannot be considered rigorously in this work. A similar restriction applied to earlier contributions on fluid flow (14) and mixing (15). It should be noted, however, that the fluid shear rates in the region immediately before a heat exchanger are usually at a high level, particularly if a pump is located just upstream from the heat exchanger. Since this represents usual industrial practice, thixotropic and rheo-

# Infinite Pseudoplasticity

As the flow-behavior index n' decreases toward zero the velocity profile of a fluid flowing through a round tube progressively flattens until a perfectly flat profile or plug flow is reached at the limit of n' equal to zero (13, 22). A solution of the Fourier-Poisson heatconduction equation for this limit was made by Graetz (7) and was reviewed in detail by Drew (6). This solution, shown as curve C of Figure 2, becomes unwieldy for values of the Graetz number  $(wC_p/kL)$ above about 500; however, for values of  $wC_n/kL$  above 100 an asymptotic solution extending the one given by Boussinesq (3) is possible (22). This solution may be

$$\frac{h_a D}{k} = \frac{8}{\pi} + \frac{4}{\pi} \left( \frac{wC_p}{kL} \right)^{1/2} \tag{7}$$

and is shown as line B on Figure 2.

For values of  $wC_p/kL$  below about 5 the theoretical curves for plug flow approach the limiting case of an outlet fluid temperature equal to the wall temperature of the tube. This limit is shown as curve A of Figure 2 and may be described (12) by the heat-balance equation:

$$\frac{h_a D}{k} = \frac{2}{\pi} \frac{w C_p}{k L} \tag{8}$$

# Infinite Dilatancy

For this case  $(n' = \infty)$  the velocity profile of a fluid flowing in a round tube is conical (13, 22). This velocity profile accordingly is especially well suited to the use of a Leveque-type approximation, as Equation (1) is obeyed at all radii. Placing  $n' = \infty$  into Equation (6) and substituting the result into Equation (3) gives

$$\frac{h_a D}{k} = 1.590 \left(\frac{wC_p}{kL}\right)^{1/3} \tag{9}$$

This equation is shown as curve F of Figure 2.

### **Newtonian Behavior**

A theory of heat transfer to Newtonian fluids in laminar flow was first given by Graetz (8). The assumptions upon which his solution is based, as well as the solution itself, are adequately presented by Drew (6), Jakob (10) and McAdams (12) and need not be repeated here. The resulting equation is plotted as curve E in Figure 2. Below Graetz numbers of about 3 the results are identical with curve A; i.e., the exit-fluid temperature approaches the wall temperature very closely.

Generally speaking, one is interested in high mass flow rates and relatively short tubes, i.e., in high Graetz numbers. For Graetz numbers above about 100 the Graetz equations become unwieldy. Levegue (11) noted this deficiency and proceeded to develop his asymptotic equation. An outline of the development and a thorough review is given by reference 6. Leveque's equation describing the transfer of heat to a Newtonian fluid flowing in streamline motion through round tubes is shown as curve D of Figure 2 and may be obtained by placing  $\alpha = 8V/D$  in Equation (4) and substituing the result into Equation (3):

$$\frac{h_a D}{k} = 1.75 \left(\frac{wC_p}{kL}\right)^{1/3} \tag{10}$$

The equation most recently recommended by McAdams (12) for this situation (high Graetz number, laminar flow) is

$$\frac{h_a D}{k} \left(\frac{\mu_w}{\mu}\right)^{0.14} = 1.75 \left[\frac{wC_p}{kL} + 0.04 \left(\frac{D}{L} N_{G_r} N_{P_r}\right)^{0.75}\right]^{1/3}$$
(11)

Its similarity to the above Leveque equation is obvious. The viscosity-ratio term represents the familiar Sieder-Tate correction factor to account for the distortion of the assumed velocity distribution by the radial temperature gradient. Presumably a similar correction factor will be required in all the non-Newtonian work; this will be discussed later. The last group of terms on the right-hand side of Equation (11) corrects for changes in heat transfer rate due to natural convection currents caused by the temperature gradients. For the viscous gels studied in the present work, as with most non-Newtonian fluids likely to be processed in the laminar region, this term may be assumed negligible.

Pigford (19) has recently developed a series of equations which theoretically account for the effects of both natural convection and radial viscosity variations (due to the temperature gradient) upon the Newtonian heat transfer rate. The simpler empirical approach of the Sieder-Tate correction factor was used in this work, however, to avoid the mathematical complications due to these factors, and

the complex problem of non-Newtonian behavior were considered theoretically.

In summary of this section, the limiting cases of non-Newtonian behavior have been defined and the solution to the heat transfer problem corresponding to each has been given. A brief discussion was also devoted to the theory of heat transfer to Newtonian fluids as Newtonian behavior may be thought of as a special intermediate case of non-Newtonian behavior. The results are summarized in Figure 2. Heat transfer coefficients for all pseudoplastic and Bingham plastic non-Newtonian fluids should lie between the curves for a parabolic velocity distribution (AED) and those for uniform velocity distribution, or plug flow (curves ACB). Similarly, all coefficients for dilatant fluids should lie between curves AED for a parabolic velocity distribution and curve F for a conical velocity distribution. The remainder of this theoretical section will be devoted to finding methods of interpolating between

Inspection of these equations (or of curves D and F of Figure 2) shows that even in the case of infinite dilatancy  $n'=\infty$ ) the heat transfer coefficient is still 90.9% of that for a Newtonian fluid at the same Graetz number. Experimental verification of Equation (12) for fluids of reasonable dilatancy (n' in the neighborhood of 2.0) is therefore probably impractical since the difference between the results for Newtonian and dilatant fluids is within the usual experimental error.

Below Graetz numbers of 20 an even closer similarity between heat transfer rates for Newtonian and dilatant fluids may be expected to exist. For this reason, the tentative design procedure for dilatant fluids at all Graetz numbers may be stated as

$$\delta^{1/3} = \frac{\left(\frac{h_a D}{k}\right)_{dilatant}}{\left(\frac{h_a D}{k}\right)_{Newtonian}} = \left(\frac{3n' + 1}{4n'}\right)^{1/3}$$

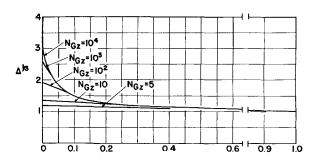


Fig. 3. Theoretical ratio of non-Newtonian to Newtonian heat transfer rates in laminarflow pseudoplastic fluids.

FLOW BEHAVIOR INDEX n'

these limiting cases for intermediate amounts of non-Newtonian behavior.

# Dilatant Fluids (1.00 $< n' < \infty$ )

Above a Graetz number of about 20 the Leveque approximation may be used to predict heat transfer rates for both Newtonian fluids (n' = 1.00) and infinitely dilatant fluids  $(n' = \infty)$  in laminar flow. Accordingly it is obviously a valid assumption that the same method may be used to calculate heat transfer coefficients within the entire region between these limits. From Equations (6) and (3), therefore,

$$\frac{h_a D}{k} = 1.75 \left(\frac{3n'+1}{4n'}\right)^{1/3} \left(\frac{wC_p}{kL}\right)^{1/3}$$
(12)

01

$$\frac{h_a D}{k \delta^{1/3}} = 1.75 \left(\frac{w C_p}{k L}\right)^{1/3} \tag{13}$$

where

$$\delta^{1/3} = \left(\frac{3n'+1}{4n'}\right)^{1/3} \tag{6}$$

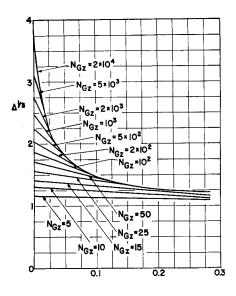
Below Graetz numbers of about 20 this procedure is not exact but will probably be slightly conservative. Above  $N_{Gz}=20$  this relationship is identical to that given by Equations (12) and (13) and hence is indicated as being theoretically exact under these conditions.

# Pseudoplastic Fluids (0 < n' < 1.00)

The problem in this case is one of interpolation between curves C-B (n'=0) on one hand and curves E-D (n'=1.00) on the other. For this purpose a correction factor  $\Delta$  may be defined as follows:

$$\Delta^{1/3} = \frac{(h_a D/k)_{non-Newtonian}}{(h_a D/k)_{Newtonian}}$$

When n' = 0,  $\Delta^{1/3}$  is given by the ratio of the ordinates of the aforementioned curves at any chosen value of the Graetz number. At values of n' close to unity, the value of  $\Delta$  will coincide with that of  $\delta$  as given by Equation (6), provided the Leveque approximation is also valid  $(N_{Gz}$  above about 100). Any method of interpolation between curves CB and ED must, therefore, satisfy the following



FLOW BEHAVIOR INDEX n1

Fig. 4. Theoretical ratio of non-Newtonian to Newtonian heat transfer rates for extremely pseudoplastic fluids in laminar flow.

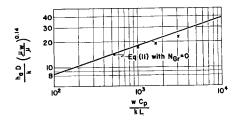


Fig. 5. Proof of equipment: comparison of Newtonian data with recommended (12) equation.

conditions: (a) the method must give the correct value of  $\Delta$  when n'=0 and (b) it must give  $\Delta=\delta$  when n' approaches unity, where Equations (13) and (6) are known to be either rigorously valid or a very close approximation  $(N_{Gz} > 100)$ 

The simplest possible interpolation procedure was arbitrarily chosen in this work; viz., a linear interpolation was made between the value of  $\Delta^{1/3}$  at n' = 0 and the point at which this straight line first became tangent to  $\delta^{1/3}$  at higher values of n' at selected values of the Graetz numbers. These interpolating curves are shown in Figures 3 and 4.

In Figure 3 the uppermost curve represents Equation (6) to an n' as low as 0.02; in Figure 4 the corresponding value of n' to which the Leveque theory is valid is about 0.01 at  $N_{Gz} = 2 \times 10^4$ .

The most significant conclusion indicated by these two figures is that at the higher Graetz numbers Equations (13) and (6) are valid to extremely low values of the flow-behavior index n'. Even at as low a value of the Graetz number as 100, the interpolation follows Equation (6) until n' drops below about 0.10. The importance of the assumed validity of a linear interpolation is therefore not critical in view of the small range of the flow-behavior index over which this is used. At lower values of the Graetz number, on the other hand, while the linear interpolation is used over a wide range of values of n' the value of  $\Delta^{1/3}$  is so near unity that the exact choice of interpolation procedures is still not critical. The fact that high Graetz numbers and values of n' above 0.10 are usual

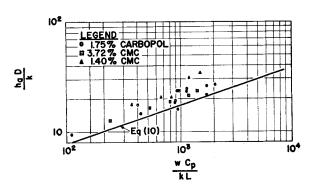


Fig. 6. Comparison of experimental non-Newtonian data with theoretical Newtonian curve.

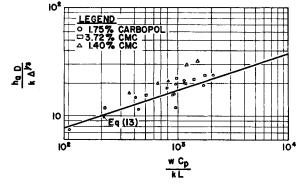


Fig. 7. Comparison of data with theoretical curve for non-Newtonian fluids.

industrial practice means that only rarely will Figures 3 and 4 be used under conditions such that Equation (6) is not applicable. The chief value of Figures (3) and (4) may therefore be considered to be that of having defined the limits within which Equation (6) is valid.

The theoretical methods of predicting heat transfer coefficients for non-Newtonian fluids in laminar flow may be summarized as follows:

(a) For  $wC_p/kL$  above 100 and n' between 0.10 and infinity,

$$\frac{h_a D}{k} = 1.75 \delta^{1/3} \left( \frac{w C_p}{k L} \right)^{1/3} \tag{13}$$

where

$$\delta = \frac{3n' + 1}{4n'} \tag{6}$$

(b) For values of n' below 0.10 at all values of  $wC_p/kL$ , and for n' between 0.10 and 1.00 when  $wC_p/kL$  is below 100,

$$\frac{(h_a D/k)_{non-Newtonian}}{(h_a D/k)_{Newtonian}} = \Delta^{1/3}$$

where  $\Delta^{1/3}$  is obtained from Figure 3 or 4. The term  $(h_a D/k)_{Newtonian}$  may be obtained from Figure 2 under all conditions.

(c) If n' is above unity and  $wC_p/kL$  below 100, procedure b is also followed, but the correction factor is taken as

$$\delta^{1/3} = \left(\frac{3n'+1}{4n'}\right)^{1/3}$$

This procedure appears to be rigorous down to values of the Graetz number of about 20.

These three alternatives encompass all ranges of Newtonian and non-Newtonian behavior considered in this paper. Most industrial problems may be expected to fall into procedure a.

As the foregoing theoretical conclusions have been based on a number of assumptions, experimental proof must be obtained to show their validity and to define the need for a Sieder-Tate type of correction factor. Since the equipment and materials used were chosen to simulate industrial practice, experimental data were obtainable to verify only the first of the three design procedures.

# EXPERIMENTAL EQUIPMENT

A gravity-fed, variable-speed Moyno "progressing-cavity" type of pump was used in this work. Upon leaving the pump the fluid passed through an orifice mixer fitted with a thermocouple for temperature equalization and measurement. The mixer was followed in turn by a 20-diam.-long calming section and a 19-ft.-long heat transfer test section. This exchanger had an inside diameter of 1.368 in. and was surrounded by 3- and 5-in.-diam. copper water tubes. The inner annulus thus formed

served as a steam jacket for the test section itself and the outer annulus acted as an isothermal insulator since the same steam was admitted to both annuli. Upon leaving the test section the fluid was passed through a second orifice mixer for measurement of the exit-fluid temperature. Heat balances were obtained to check the measured temperature rise of the fluid against the rate of steam condensation. The wall temperature of the 1.368-in. test section was measured by seven thermocouples distributed along its length. This exchanger was generally similar to that described in detail by Bonilla and coworkers (2). Auxiliary equipment was conventional.

To measure the isothermal-fluid-flow properties, both a Stormer concentriccylinder viscometer and a capillary-tube viscometer were used; both were calibrated with National Bureau of Standards oils. The capillary-tube viscometer was equipped with a constant-temperature bath to allow operation at elevated temperatures. Operating pressures were supplied by compressed nitrogen controlled by a Grove regulator and measured by calibrated gauges. laboratory-test pressure The Stormer viscometer was used primarily for purposes of comparison with the capillaryviscometer room-temperature results. Methods of converting these experimental viscometric data to the rheological parameters K' (or  $\gamma$ ) and n' have been described elsewhere (13, 15).

The thermal conductivities of the fluids were measured by the steady state method with two chambers in series. The upper chamber contained the fluid of unknown conductivity and the lower a fluid of known conductivity (water). A thermocouple at the center of the plate below and above each cell indicated the surface temperature there. A constant-temperature bath above the two chambers supplied the heat flux which passed through the two chambers to an ice-water bath below. Downward heating was used to minimize convection effects. The entire apparatus was insulated and could easily be disassembled.

Fluid heat capacities were measured by adding a known amount of electrical energy to an insulated, agitated bath containing the fluid of unknown heat capacity in six 1-in.-diam. by 10-in.-long sample tubes. The rise in the temperature of the entire bath was measured by a calibrated thermometer and the heat capacity of the fluid calculated after subtracting the capacity of the bath (other than the sample fluid) as determined by calibration with water and checked with benzene.

Fluid densities were measured by a modified pycnometric method. Full experimental details are available (22).

# RESULTS

Proof of the absence of unusual experimental conditions and of any calculational or procedural errors was obtained by first taking heat transfer data on molasses, a Newtonian fluid. Comparison of the experimental results with Equation (11) is shown in Figure 5. The maximum deviation of an experimental data point from the curve was -10%.

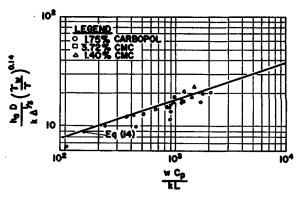


Fig. 8. Comparison of data with theoretical relationship after introduction of the Sieder-Tate correction factor.

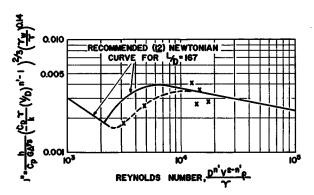


Fig. 9. Non-Newtonian heat transfer in the transition and turbulent-flow regions.

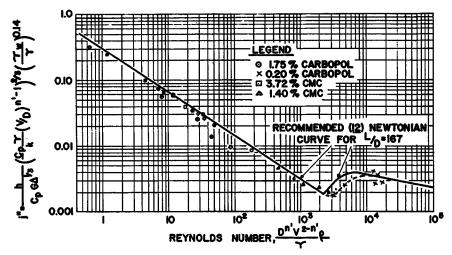


Fig. 10. Non-Newtonian heat transfer: summary and correlation of all data.

In view of the fact that maximum deviation of prior-art data was  $\pm 30\%$  (12), it was concluded that all procedural details of the present work were entirely satisfactory.

# Heat Transfer to Non-Newtonian Fluids:

Heat transfer data were taken on the following three non-Newtonian fluids: (1) a 1.75% solution of Carbopol in

water (the flow behavior index n' of this fluid varying between 0.18 and 0.40, depending upon the temperature and shear rate), (2) a 3.72% solution of sodium carboxymethylcellulose (CMC) in water (n') varying between 0.43 and 0.51 for this fluid), and (3) a 1.40% CMC solution with a flow-behavior index of 0.70.

A plot of the original data on these fluids is shown in Figure 6. Since no factor is included here to correct for the

deviation of the fluids from Newtonian behavior, the data fall, as would be expected, considerably above the curve. Introduction of the correction factor  $\Delta^{1/s}$  (=  $\delta^{1/s}$ ), as in Figure 7, brings the data more nearly in line with the theoretical curve. There is still much scatter, however, the maximum deviations being +35 and -30%.

It is instructive at this point to compare these results with those reported for Newtonian fluids. McAdams (12), in reviewing the Newtonian data, pointed out that the scatter in that case is as great as  $\pm 100\%$ . The causes of this great scatter are the distortion of the velocity profile and the free-convection effects resulting from the temperature gradient in the fluid. For fluids of high consistency, which certainly includes these highly non-Newtonian fluids, free convection effects are very small and may be neglected. The distortion of the velocity profile for Newtonian fluids is usually allowed for empirically by introduction of the Sieder-Tate viscosity ratio:  $(\mu/\mu_w)^{0.14}$ .

As will be shown later, the flow properties of the 1.75% Carbopol-water system were least dependent on temperature and the 3.72% and 1.4% CMC-water systems have an increasing dependence of the flow properties on temperature in the order given. Inspection of Figure 7 shows the trend of data away from the curve to be in the same order. These facts suggest that a correction factor analogous to the viscosity ratio  $(\mu/\mu_m)$ would aid in correlation of the data. The denominator  $\gamma$  of the generalized Reynolds number seems a plausible substitute for the viscosity of Newtonian fluids, as it uniquely defines the consistency of non-Newtonian fluids of any given flow-behavior index.

The data including the empirical correction factor  $(\gamma/\gamma_w)^{0.14}$  are plotted in Figure 8. With the exception of a few of the points for the Carbopol-water system, this empirical correction factor adequately correlates the experimental results. Thus the final correlating equation (for  $wC_p/kL$  above 100) adopted in this work for both Newtonian and non-Newtonian fluids is

$$\frac{h_a D}{k \Delta^{1/3}} = 1.75 \left(\frac{w C_p}{k L}\right)^{1/3} \left(\frac{\gamma}{\gamma_w}\right)^{0.14} \quad (14)$$

For  $wC_v/kL$  below 100, the procedures previously outlined are recommended upon introduction of the  $(\gamma/\gamma_w)^{0.14}$  term to account for distortion of the theoretically assumed velocity profile.

The significance of each of the non-Newtonian terms in Equation (14) has already been discussed. For Newtonian fluids,  $\Delta^{1/3} = 1.00$ ,  $\gamma$  reduces to  $\mu$  and  $\gamma_w$  to  $\mu_w$ . Accordingly the foregoing equation reduces to the one usually recommended for this special case when natural-convection effects are absent.

The mean deviation of the non-Newtonian data from Equation (14) is 13.5%. The two points furthest from the curve were for those runs having the greatest error in the heat balances; hence the presence of some experimental errors is indicated. The fact that most of the

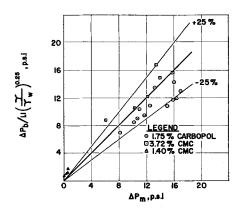


Fig. 11. Comparison of corrected bulktemperature pressure drop with measured

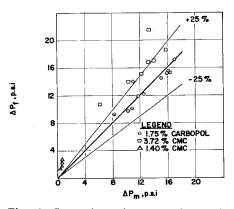


Fig. 12. Comparison of pressure drops calculated at the film temperature with measured values.

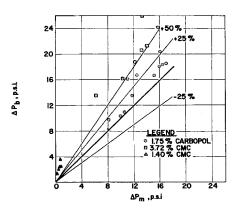


Fig. 13. Comparison of pressure drop calculated for isothermal conditions with nonisothermal measured values.

data points fall below the curve may be related to the fact that all heat transfer coefficients were based upon the input heat flux as measured by the steam-condensation rate. This was considered to be the most direst, hence accurate, procedure. Nevertheless, these values were usually somewhat lower than those calculated from the temperature rise of the fluid. In any case, all deviations are within experimental error and no adjustment of the curve is felt warranted on the basis of these data alone.

# Heat Transfer Outside the Laminar Flow Region

It has been shown that the conventional Newtonian-friction-factor-Reynolds-number curve is useful for interpretation of non-Newtonian data upon generalization of the Reynolds number (14). More recently the utility of this generalized Reynolds number has been shown to extend to the problem of mixing-power requirements (15) as well. Its use in correlation of non-Newtonian heat transfer data is therefore suggested by the close analogy between these three problems for Newtonian fluids.

For the case of Newtonian heat transfer, the relevant dimensionless groups are the Reynolds number  $(DV\rho/\mu)$ , the Prandtl number,  $C_p\mu/k$ , and either the Nusselt (hD/k) or Stanton  $(h/C_pG)$  numbers. In addition the Sieder-Tate  $(\mu/\mu_w)^{0.14}$  factor is required. Finally, the L/D ratio is important if data in the laminar and transition regions are considered.

Generalization of the foregoing dimensionless groups for applicability to non-Newtonian as well as to Newtonian fluids involves, in addition to the Sieder-Tate factor already considered, the Prandtl and Reynolds numbers—as both contain the Newtonian viscosity. Unquestionably, both may be used in their present form if a judiciously chosen apparent viscosity  $\mu_a$  is used to replace the true Newtonian viscosity. A possible method of determining this apparent viscosity is to equate the Newtonian Reynolds number to the generalized one, as follows:

Ιf

$$\frac{DV\rho}{\mu_a} = \frac{D^{n'}V^{2-n'}\rho}{\gamma}$$

then

$$\mu_a = \gamma (V/D)^{n'-1}$$

Substitution into the conventional Prandtl number gives

$$\frac{C_{p}\mu_{a}}{k} = \frac{C_{p}\gamma}{k} \left(V/D\right)^{n'-1}$$

Accordingly, the relevant non-Newtonian dimensionless groups become

the generalized Reynolds number:

$$\frac{D^{n'}V^{2-n'}\rho}{\gamma}$$

the generalized Prandtl number:

$$\frac{C_{p}\gamma}{k}\left(V/D\right)^{n'-1}$$

either the Nusselt or Stanton numbers:

$$\frac{hD}{k}$$
 or  $\frac{h}{C_{v}G}$ 

the dimensionless consistency ratio:

$$\frac{\gamma}{\gamma_w}$$

In the laminar and transition regions the L/D term as well as the correction factor  $\Delta^{1/3}$  (or  $\delta^{1/3}$ ) is also relevant. Presumably the latter would decrease in importance much as the L/D term as the flow becomes fully turbulent and the non-Newtonian fluid loses its peculiar identity.

If these generalizations are both valid and adequate, the functional relationships between the dimensionless groups must be the same as found in conventional Newtonian heat transfer. If this were not the case, the generalized correlations for non-Newtonian fluids would not reduce to the well-established Newtonian formulas upon substitution of n' = 1.00 (and  $\gamma = \mu$ ) into the generalized Reynolds and Prandtl numbers. The only expected exception to this consideration would occur in the transition region, as it has been established (14) that the transition from laminar to fully developed turbulent flow is more gradual for highly pseudoplastic fluids, for example, than for Newtonians. For a fluid with a flowbehavior index of about 0.80, these considerations indicate a transition region extending to Reynolds numbers of about 10,000 although quantitative data are not available.

In order to preview this important problem of heat transfer outside the laminar region, a small number of data were taken with a dilute (0.2%) Carbopol solution having a flow-behavior index varying between 0.74 and 0.82. These data are shown in Figure 9 on a conventional j-factor-Reynolds-number plot using the aforementioned dimensionless groups. The solid line represents the conventional Newtonian relationship recommended by McAdams (12) and the dashed line is intended to represent a curve through the data points.

It is seen that the experimental data are in perfect confirmation of all the expected trends. The experimental errors (leading to a scatter of  $\pm 20\%$ ) and the paucity of data, however, prevent any very firm conclusions. In particular, the flow-behavior index of the fluid was sufficiently high that the non-Newtonian correction factor  $\Delta^{1/3}$  (Figure 3) could be taken equal to unity within experimental error; hence no information is available on its importance outside the laminar region.

Figure 10 summarizes all the experimental data on the conventional j-factor type of plot. In view of the good agreement between the theoretical analysis and the experimental data in the laminar region, heat transfer estimates may be made with considerable certainty here. Outside the laminar region the present results may be considered indicative primarily of a method of approach, which may be either refuted or confirmed by more extensive work. Up to Reynolds numbers of 15,000 to 20,000 Figure 10 may be used for order-of-magnitude estimates, however, and as such represents some improvement on the prior art.

## Nonisothermal-pressure-drop Calculations

McAdams (12) suggests two methods for correlation of nonisothermal–pressure-drop data for Newtonian fluids.

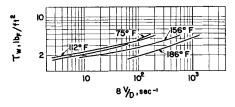


Fig. 14. Rheological properties of the 1.75% Carbopol-water system.

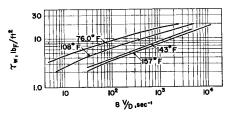


Fig. 15. Rheological properties of aqueous 3.72% CMC (sodium carboxymethylcellulose) solution.

The most recent edition of this book recommends that the actual pressure drop may be calculated from the usual laminar-flow equations upon substitution of the fluid viscosity at a film temperature defined by

$$t_f = t_b + \frac{t_w - t_b}{4}$$
 (15)

The older second edition of this book recommended either use of this procedure or consideration of the pressure drop calculated at the bulk temperature as the product of the nonisothermal pressure drop and the factor 1.1  $(\mu/\mu_w)^{0.25}$ .

The results shown in Table 1 for the Newtonian fluid, molasses, indicate that neither method predicts the nonisothermal pressure drop very accurately, and these data accordingly leave little basis for choice of one method over the other.

For non-Newtonian fluids, the method corresponding to the first of these approaches involves evaluating the fluid consistency,  $\gamma$  and the flow-behavior index n' at the film temperature defined by Equation (15). These are then used with the generalized Reynolds number to calculate the friction factor or may be substituted directly in the generalized equivalent of Poiseuille's law (14):

Table 1.

Nonisothermal Pressure-drop Data
for Molasses

Calculated pressure drop, lb./sq. in., with

Velocity, ft./sec.		evaluated at film	Bulk viscosity divided by $1.1(\mu/\mu_w)^{0.28}$
3.22 $1.90$ $5.82$	$egin{array}{c} 2.6 \\ 0.5 \\ 4.5 \\ \end{array}$	$2.2 \\ 1.0 \\ 3.3$	$egin{array}{c} 2.4 \\ 1.2 \\ 3.4 \end{array}$

I.D. of pipe: 1.368 in.

Distance between pressure taps: 19.07 ft.

$$\Delta P = \frac{32\gamma L V^{n'}}{g_c D^{n'+1}} \tag{16}$$

McAdams's older recommendation, involving the one-quarter power of the viscosity ratio, was adopted by calculating the bulk-temperature pressure drop by means of Equation (16) and dividing the result by

$$1.1 \left(\frac{\gamma}{\gamma_w}\right)^{0.25}$$

Figure 11 compares the results obtained upon calculating the nonisothermal pressure drop in the last manner with the actual measured pressure drop. Figure 12 shows a similar comparison for the case in which the calculated pressure drop was based upon Equations (15) and (16). The improvement obtained by either method may be noted by comparison with Figure 13, in which the calculated pressure drop was obtained by use of bulk-temperature physical properties. If one neglects the 1.4% CMC data, for which the measured pressure drops were too small to be determined accurately, all but five data points are within 25% of the 45° line of equivalence in both Figures 11 and 12. The mean deviation of the data from the line of equivalence is 14% in Figure 11 and 18% in Figure 12. Thus there is very little basis for choice of one method over the other; in general the data are overcorrected in Figure 11 and undercorrected in Figure 12. Fortunately the  $\pm 25\%$  uncertainty in use of these methods, although large, is frequently not prohibitive.

As the problem of nonlaminar fluid pressure drop is not yet entirely clear for non-Newtonian fluids flowing isothermally, it is not fruitful to discuss noniso-

thermal flow outside the laminar region at this time.

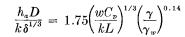
# Rheological Properties of Fluids Used

The fundamental flow groups for interpretation of the capillary-viscometer data,  $D\Delta p/4L$  and 8V/D, are plotted in Figures 14 to 17 for the four fluids used. To avoid confusion, the experimental

i.e. the liquid was slightly non-Newtonian at this temperature. At the higher temperatures used in the heat exchanger, however, no non-Newtonian behavior could be detected.

# Other Physical Properties

The thermal conductivities of the four non-Newtonian fluids used ranged from



where

$$\delta = \frac{3n' + 1}{4n'}$$

As shown by Figures 8 and 10, this equation has been checked with three non-Newtonian fluids over the following ranges of variables:

$$n': 0.18 \text{ to } 0.70$$

$$\frac{wC_{p}}{kL}$$
: 100 to 2050

$$N_{Re}: 0.65 \text{ to } 2100$$

The Newtonian equation recommended by McAdams (12) represents the special case obtained by substitution of n' = 1.00

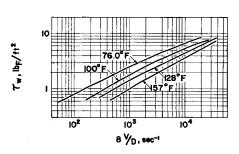


Fig. 16. Rheological properties of aqueous 1.40% CMC solution.

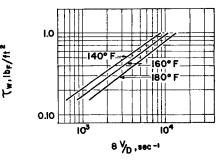


Fig. 17. Rheological properties of aqueous 0.20% Carbopol solution.

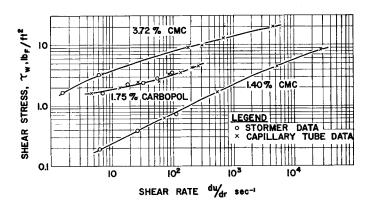


Fig. 18. Equivalence of rheological data as obtained with rotational and capillary-tube viscometers.

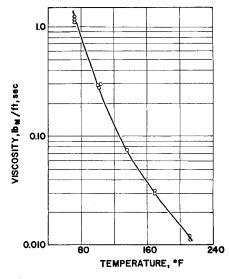


Fig. 19. Viscosity—temperature relationship for molasses.

data points have been omitted. However, the individual curves are well supported in that the maximum deviation from any curve was only 14% and the mean deviation was less than 2%. On these logarithmic plots the slope of the tangent to the curve gives the value of the flow-behavior index n' at the particular value of 8V/D chosen.

In order to show the validity of the capillary-tube data, the 75° to 76°F. capillary-tube data for the three viscous fluids were compared with similar shear-stress-shear-rate data measured with a Stormer rotational viscometer by means of the usual relationships (13) for calculation of the shear rates. As Figure 18 shows, the agreement between the two viscometers is excellent.

Experimental viscosity data for the molasses used are shown in Figure 19. The three points shown at 72.5°F. correspond to three different shear rates;

the conductivity of water to as much as 25% below water. The heat capacities for these same fluids were found to be within 3% of the value for water at the same temperature. Since this was approximately equal to the reliability of the measurements, the heat capacity of water was used in the calculations. Similarly, the densities of the non-Newtonian fluids were found to be equal to the density of water.

# SUMMARY

The recommended design procedure for calculation of laminar-region heat transfer coefficients for fluids in which natural-convection effects are not present may be summarized as follows:

1. For most fluids likely to be encountered (n' above 0.10) at reasonably high flow rates ( $wC_p/kL$  above 100)

 $(\delta = 1.00 \text{ and } \gamma = \mu)$  into the foregoing equation. The mean deviation of the non-Newtonian data from this equation was 13.5% and the maximum deviation 30%. These figures compare very favorably with those usually reported for Newtonian fluids.

2. For extremely highly pseudoplastic fluids (n' below 0.10) and for all pseudoplastics at low flow rates ( $wC_p/kL$  below 100), the ordinate of curve AED of Figure 2 at any given value of the Graetz number is taken as equal to  $[(k_aD)/(k\Delta^{1/3})](\gamma_w/\gamma)^{0.14}$ . The term  $\Delta^{1/3}$  is evaluated from Figure 3 or 4.

3. For dilatant fluids the ordinate as read from curve AED of Figure 2 is equated to the quantity  $[(h_n D)/(k\delta^{1/3})]$   $(\gamma_w/\gamma)^{0.14}$ , where  $\delta = 3n' + 1/4n'$ . At higher flow rates  $(wC_p/kL)$  above 100) this procedure is identical to that of part 1.

The procedures outlined under 2 and 3 have not been verified experimentally

but may be recommended as being at least good approximations in view of the excellent agreement between theory and experiment found in part 1. It should be noted that pseudoplastic and Binghamplastic fluids have higher transfer coefficients in the laminar region than do Newtonians. The reverse is true for dilatant materials, for which, as a matter of fact, the foregoing recommendations predict heat transfer coefficients which are always within 10% of those for Newtonian fluids.

Heat transfer rates outside the laminarflow region may be estimated from Figures 9 and 10. As the importance of the term  $\Delta^{1/3}$  has not been established outside the laminar region, the conservative procedure of assuming it to be equal to unity for pseudoplastic fluids is recommended.

The usual Newtonian methods for  $nonisothermal-pressure-drop\ calculations\ in$ laminar flow may be applied to the non-Newtonian fluids studied with an accuracy of about  $\pm 25\%$ . No attempt has been made to predict nonisothermal pressure drops outside the laminar region.

Attention must be drawn to the strong need for additional data for all the problems discussed. In particular, the limitations of the present work are that only one tube size and length has been studied and all the data have been for heating of the fluids.

# **ACKNOWLEDGMENT**

This work was started on funds provided by a duPont Fellowship and continued under sponsorship of the Office of Ordnance Research, U. S. Army. The Continental-Diamond Fibre Division of The Budd Company made available the Stormer viscometer used.

# NOTATION

- = fluid heat capacity, B.t.u./  $(lb_{M})(^{\circ}F.)$
- du/dr = velocity gradient or shear rate,  $\sec^{-1} (du/dr)_i$  refers to the shear rate at the (inner) bob of a viscometer and  $(du/dr)_w$  to the shear rate at the wall of a tube
- D= tube diameter, ft.
- = acceleration of gravity, ft./sec.
- = dimensional conversion factor,  $g_c$  $32.2 \text{ (ft.)(lb.}_{M})/(\text{sec.}^{2})(\text{lb.}_{F}.)$
- mass velocity,  $lb._M/(hr.)$ (sq. ft.) Gor lb.<sub>M</sub>/(sec.)(sq. ft.)  $G = V\rho$
- hheat transfer film coefficient, B.t.u./(hr.)(sq. ft./°F.)
- a film coefficient based upon an  $h_a$ arithmetic-mean temperature difference
- j''= grouping of dimensionless terms:

$$j^{\prime\prime} = \frac{h}{C_{\rm p} G \Delta^{1/3}}$$

- = thermal conductivity, B.t.u./ (hr.)(ft.)(°F.)
- = fluid-consistency index,  $(lb._F)$ (sec.")/sq. ft. If the fluid happens to be Newtonian  $K' = \mu/g_c$
- L= tube length, ft.
  - flow-behavior index, dimensionless. The numerical value of n'lies between zero and unity for pseudoplastic and Binghamplastic fluids, is equal to unity for Newtonians, and is greater than unity for dilatant fluids. The flow-behavior index at a given shear stress is equal to the slope of the logarithmic curves of Figures 14 to 17
- $\Delta P$ = pressure drop,  $(lb._F)/(sq. ft.)$  or  $(lb._F)/(sq. in.)$ .  $\Delta P_b$  refers to a pressure drop calculated at the fluid bulk temperature,  $\Delta P_f$  to that calculated at the film temperature, and  $\Delta P_m$  to the measured pressure drop
- = radial distance, ft.
- = radius of tube, ft.
  - temperature,  ${}^{\circ}F$ . ( $t_b$  denotes the fluid bulk temperature,  $t_f$  the film temperature, and  $t_w$  the temperature at the wall.)
- $\Delta t$ temperature difference or driving force, °F.
- local velocity, ft./sec.
- Vaverage or bulk velocity, ft./sec.
- = mass flow rate,  $lb_{M}/hr$ . or w $lb._{M}/sec.$
- velocity gradient at wall of tube, sec.-1
- coefficient of expansion, °F.-1
- = fluid consistency,  $lb._{M}/(ft.)$ (sec.<sup>2-n'</sup>.)  $\gamma_w$  refers to the term evaluated at the wall temperature.  $\gamma = g_c K' 8^{n'-1}$ . For a Newtonian fluid,  $\gamma = g_c K' = \mu$ .
- δ = ratio of non-Newtonian to Newtonian shear rates,  $\delta = \alpha/8V/D$ = (3n' + 1)/4n' dimensionless
- $\Delta^{1/3}$ = ratio of non-Newtonian to Newtonian heat transfer rates, didimensionless. In regions for which the Leveque theory is valid,  $\Delta^{1/3} = \delta^{1/3}$ .
- = Newtonian viscosity,  $lb_{M}/(sec.)$ (ft.).  $\mu_w$  denotes viscosity evaluated at the wall temperature and  $\mu_a$  the "apparent" viscosity of a non-Newtonian fluid.
- = 3.14 . . .

ρ

- = density,  $lb._{M}/cu.$  ft.
- = shear stress, lb.<sub>F</sub>/sq. ft.  $\tau_w$  denotes the shear stress at the wall of a tube and  $\tau_i$  that at a viscometer bob.
- yield value of a Bingham-plastic fluid,  $lb._F/sq.$  ft.

# Dimensionless Groups

 $N_{Gr} = \text{Grashof number, } (\beta \Delta t D^3 \rho^2 g)/\mu^2$ = Graetz number,  $wC_n/kL$ 

- $\cdot \left[\frac{C_{r}\gamma}{k}(V/D)^{n'-1}\right]^{2/3} \left(\frac{\gamma_{w}}{\gamma}\right)^{0.14} \quad N_{Nu} \quad = \text{Nusselt number, taken as} \\ \quad hD/k\Delta^{1/3} \quad \text{or} \quad hD/k\delta^{1/3} \quad \text{in the}$ laminar region. For Newtonian fluids  $\Delta^{1/3} = \delta^{1/3} = 1.00$ ; hence  $N_{Nu} = hD/k$ .
  - = generalized Prandtl number,  $N_{Pr}$  $(C_p \gamma/k) (V/D)^{n'-1}$ . For Newtonian fluids n' = 1.00 and  $\dot{\gamma} = \mu$ ; hence  $N_{Pr} = C_p \mu/k$ .
  - $N_{Re}$ generalized Reynolds number,  $(D^{n'}V^{2-n'}\rho/\gamma)$ . For Newtonians, this reduces to  $DV\rho/\mu$ .
  - $N_{St}$ = Stanton number, taken as  $h/(C_{\nu}G\Delta^{1/3})$  in the laminar re-

# LITERATURE CITED

- 1. Alves, G. E., D. F. Boucher, and R. L. Pigford, Chem. Eng. Progr., 48, 385
- Bonilla, C. F., Armando Cervi, Jr., T. J. Colvin, Jr., and S. J. Wang, Chem. Eng. Progr. Symposium Ser.
- No. 5, 49, 127 (1953).

  3. Boussinesq. J., "Théorie Mathématique de la Chaleur," Bachelier, Paris (1835). See also reference 6.
- 4. Branch, R. E., M. Ch. E. thesis, Univ. Delaware, Newark (1954) (described in reference 13).
- Chu, J. C., Frank Brown, and K. G. Burridge, Ind. Eng. Chem., 45, 1686
- 6. Drew, T. B., Trans. A.I.Ch.E., 26, 26 (1931).
- 7. Graetz, L., Ann. Physik, 18, 79 (1883).
- 8. Ibid., 25, 337 (1885).
- Hedstrom, B. O. A., Ind. Eng. Chem., 44, 651 (1952).
- 10. Jakob, Max, "Heat Transfer," Vol. 1, John Wiley and Sons, Inc., New York (1949).
- 11. Leveque, M. A., Ann. Mines, 13, 201 (1928).
- 12. McAdams, W. H., "Heat Transmission," McGraw-Hill Book Company, sion," McGraw-Hill Book Company, Inc., New York (2 ed.: 1942, 3 ed.: 1954).
- Metzner, A. B., in "Advances in Chemical Engineering," Vol. I, T. B. Drew, ed., Academic Press, Inc., New York (1956).
- -, and J. C. Reed, A.I.Ch.E.Journal, 1, 434 (1955).
- 15. Metzner, A. B., and R. E. Otto, ibid., **3**, 3 (1957).
- Miller, A. P., Jr., Ph.D. thesis, Univ. Washington, Seattle (1953).
- 17. Mooney, Melvin, J. Rheology, 2, 210 (1931)
- 18. Orr, Clyde, Jr., and J. M. DallaValle, Chem. Eng. Progr. Symposium Ser. No. 9, 50, 29 (1954).
- 19. Pigford, R. L., ibid., No. 17, 51, 79
- 20. Rabinowitsch, B., Z. physik. Chem., A145, 1 (1929).
- 21. Salamone, J. J., and Morris Newman,
- Ind. Eng. Chem., 47, 283 (1955). Vaughn, R. D., Ph.D. thesis, Univ. Delaware, Newark (1956).
- Winding, C. C., F. W. Dittman, and W. L. Kranich, "Thermal Properties of Synthetic Rubber Latices," Report to Office of Rubber Reserve, Univ., Ithaca, N. Y. (1944). Cornell

Presented at A.I.Ch.E. Los Angeles meeting