Distribution of Temperature in 1-2— and 1-4— pass Heat Exchangers

Vertical Units in Which the Shell-side Pure Fluid Changes Phase and Temperature

HERBERT T. BATES

University of Nebraska, Lincoln, Nebraska

Because of practical construction advantages, heat exchangers with two or more tube passes are often used as condensers. In such cases if the exchanger is overdesigned or underloaded or if the vapor supplied is superheated, the unit may serve as both a condenser and a cooler. It is customary to ignore the temperature change of the condensing fluid when the mean temperature is calculated, provided that the cooling load is a small part of the total heat transfer load. However, when the cooling load is appreciable, this simplified approach is inaccurate.

A heat exchanger mounted vertically with the vapor entering the shell side at the top and flowing downward will be considered. If the vapor is superheated, its temperature must be reduced to the saturation value. When the vapor is desuperheated by heat transfer alone, the tube-wall temperature would have to be above the saturation temperature of the steam in order to have a dry tube wall. This might be so for the hot tube pass, but the cold tube pass would probably be wet. With water vapor the latent heat is so large compared with the sensible heat of desuperheating that revaporization of condensate would quickly remove the superheat. However, many organic vapors have latent heats low enough so that desuperheating would be accomplished primarily by heat transfer with the superheated vapor. At some cross section the vapor in the shell would become saturated and the remaining heat transfer would result in condensation. Owing to the vertical arrangement the condensate would drain to the bottom of the exchanger.

A similar case results when saturated vapor enters the top of the shell of a vertical exchanger at a rate so low that condensation does not require all the heat transfer surface. If there is very little noncondensable gas, the vacuum produced by condensation holds up the liquid in the bottom of the condenser shell and it becomes subcooled before emerging.

In either case the condensation and the cooling occur in separate sections of the exchanger. Each section has its own over-all coefficient of heat transfer, and ordinarily they are not the same because condensation of pure vapors occurs at a higher film coefficient than does cooling of either vapor or liquid.

The division between the sections should be fairly sharp in order to comply with the assumption made in the derivation to be given. This is true if the shell-side fluid is reasonably pure or if it is a constant-boiling mixture and if there is a reasonably high pressure drop through the shell.

The location of the division between the sections usually cannot be determined by test, nor does it remain fixed when the load changes. The areas required for each of the sections can be calculated, and from them the location of the division between the sections can be obtained.

In the carrying out of the derivations the following assumptions were made.

- 1. The shell-side fluid is a single pure substance or a constant-boiling mixture.
- 2. The shell-side fluid is completely mixed at each cross section but not in the direction of the tubes.
- 3. The two over-all coefficients are constant throughout their respective sections of the exchanger.
- 4. The mass rates of flow and the heat capacities of each fluid are constant.
- 5. Each tube pass contains the same transfer area.
 - 6. Heat losses are negligible.

The first two of these assumptions are necessary in order that there will be a sharp separation between the two sections of the exchanger. If a considerable amount of another substance is present in the shell-side fluid stream, the saturation temperature varies and the two sections of the exchanger become almost indistinguishable.

The equation for use with a 1-2-pass heat exchanger involving no change of phase was derived by Underwood (2) many years ago. Kern (1) made use of the same equation for situations involving change of both temperature and phase when the heating or cooling section is next to the tube return header. In these cases one of the internal tube-side

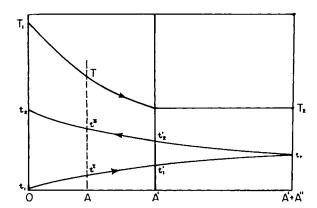


Fig. 1. Desuperheater condenser.

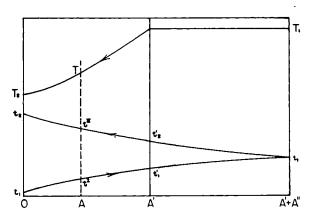
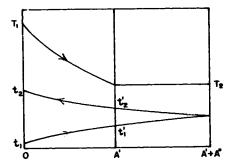
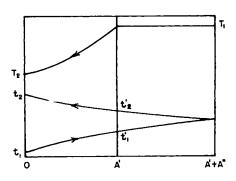


Fig. 2. Condenser subcooler.



T₁
T₂
T₃
t₄
O A' A'+A'

Fig. 3. Desuperheater-condenser heater



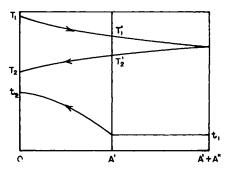


Fig. 4. Condenser-subcooler boiler superheater.

temperatures must be determined by solving a quadratic equation. The other internal temperature can be obtained by a heat balance over either section.

If the heating or cooling section is next to the channel end, i.e., the end where the tube-side fluid enters and leaves, new equations must be derived. When this is done, a new parameter M must be included which represents the ratio of the condensing load to the total heat transfer load of the exchanger.

DERIVATION OF THE EQUATIONS FOR THE 1-2-PASS DESUPERHEATER CONDENSER OF FIGURE 1

Heat Balances

Differential

 $-WC dT = wc dt^{\mathsf{I}} - wc dt^{\mathsf{II}} \quad (1)$

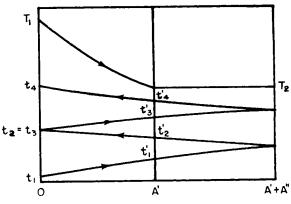


Fig. 5. Desuperheater condenser.

Condensing section

$$W\lambda = wc(t_2' - t_1')$$

Solving (10) by standard methods gives

$$(2) T = T_2 - W\lambda/WC + G_1 \exp m_1 A$$

Over-all

$$WC(T_1 - T_2) + W\lambda = wc(t_2 - t_1)$$

 $+G_2 \exp m_2 A$ (11) where

$$R = wc/WC \tag{17}$$

Around the right end

$$WC(T - T_2) + W\lambda = wc(t^{II} - t^{I})$$
(4)

$$m_1 = -\frac{U'}{2wc}(R + \sqrt{R^2 + 1})$$
 (12)

$$R(2T_1 - t_1 - t_2)$$
= $G_1(R + \sqrt{R^2 + 1})$

Substitute (12), (13), and (17) into (16)

$$WC(T-T_2) + W\lambda = wc(t^{11}-t^1)$$
(4)

$$m_2 = -\frac{U'}{2wc}(R - \sqrt{R^2 + 1})$$
 (13)

$$+ G_2(R - \sqrt{R^2 + 1})$$
 (18)

Rate Equations

Pass I

Pass II

$$wc dt^{I} = U' dA/2 (T - t^{I})$$

Evaluate (11) at the left end where $A = 0, T = T_1$

Combine (3) and (17)

$$A = 0, T = T_1$$

$$T_1 - T_2 + W\lambda/WC = R(t_2 - t_1)$$
(19)
$$T_1 - T_2 + W\lambda/WC = R(t_2 - t_1)$$
(19)

Let

 $-wc dt^{II} = U' dA/2 (T - t^{II})$

Evaluate (11) at the center where
$$A = A'$$
, $T = T_2$

 $R(t_2 - t_1) = G_1 + G_2$

(20)

and eliminate
$$G_1$$
 between (18) and (20)

$$G_2 = \frac{R(t_2 - t_1)(R + \sqrt{R^2 + 1}) - R(2T_1 - t_1 - t_2)}{2\sqrt{R^2 + 1}}$$
(21)

Combine (20) and (21)

$$G_1 = -\frac{R(t_2 - t_1)(R - \sqrt{R^2 + 1}) - R(2T_1 - t_1 - t_2)}{2\sqrt{R^2 + 1}}$$
(22)

Combine (21) and (22) with (15)

$$\frac{W\lambda}{WC} = -\frac{R(t_2 - t_1)(R - \sqrt{R^2 + 1}) - R(2T_1 - t_1 - t_2)}{2\sqrt{R^2 + 1}} \exp m_1 A' + \frac{R(t_2 - t_1)(R + \sqrt{R^2 + 1}) - R(2T_1 - t_1 - t_2)}{2\sqrt{R^2 + 1}} \exp m_2 A'$$
(23)

Multiply by $2\sqrt{R^2+1} \exp{-m_2A'}$ and solve for exp $(m_1-m_2)A$

(5)

(6)

$$\exp(m_1 - m_2)A' = \frac{R(t_2 - t_1)(R + \sqrt{R^2 + 1}) - R(2T_1 - t_1 - t_2) - 2\sqrt{R^2 + 1}}{R(t_2 - t_1)(R - \sqrt{R^2 + 1}) - R(2T_1 - t_1 - t_2)} (24)$$

Substitute (5) and (6) into (1) and re-

$$\frac{dT}{dA} = -\frac{U'}{2WC} (2T - t^{I} - t^{II})$$
 (7)

$$\frac{W\lambda}{WC} = G_1 \exp m_1 A' + G_2 \exp m_2 A'$$
 Let (15)

 $S = \frac{t_2 - t_1}{T_1 - t_1}$ (25)

Subtract (6) from (5) and rearrange

$$\frac{dt^{\mathrm{I}}}{dA} + \frac{dt^{\mathrm{II}}}{dA} = \frac{U'}{2wc} \left(t^{\mathrm{II}} - t^{\mathrm{I}} \right) \quad (8) \qquad -\frac{U'}{2WC} \left(2T_1 - t_1 - t_2 \right)$$

left end where
$$A = 0$$
, $T = T_1$, $t^1 - t_1$, $t^{11} = t_2$

$$-\frac{U'}{2WC} (2T_1 - t_1 - t_2)$$

 $= G_1 m_1 + G_2 m_2 \quad (16)$

Differentiate (11) with respect to A, equate to (7), and evaluate this at the

$$M = \frac{W\lambda}{wc(t_2 - t_1)} \tag{26}$$

Divide numerator and denominator of (24) by $R(t_2 - t_1)$ and substitute (17),

Rearrange (4)

$$t^{\text{II}} - t^{\text{I}} = (WC/wc)(T - T_2) + (W\lambda/wc) \quad (9)$$

 $\exp(m_1 - m_2)A'$

$$= \frac{R + \sqrt{R^2 + 1} - (2/S - 1) - 2M\sqrt{R^2 + 1} \exp{-m_2A'}}{R - \sqrt{R^2 + 1} - (2/S - 1)}$$
(27)

Differentiate (7) with respect to A and substitute in (8) and (9).

Combine (12) and (13):

$$(m_1 - m_2)A' = -U'A'/wc\sqrt{R^2 + 1}$$
 (28)

(25), and (26) into (24):

$$\frac{d^2T}{dA^2} + \left(\frac{U'}{WC}\right)\frac{dT}{dA} - \left(\frac{U'}{2wc}\right)^2$$

Multiply numerator and denominator of
$$(27)$$
 by S , take logarithms, and substitute (28) :

$$\cdot \left(T - T_2 + \frac{W\lambda}{wc}\right) = 0 \quad (10)$$

$$\cdot \left(T - T_2 + \frac{W\lambda}{wc}\right) = 0 \quad (10) \quad \frac{U'A'}{wc} = \frac{1}{\sqrt{R^2 + 1}} \ln \frac{2 - S(R + 1 - \sqrt{R^2 + 1})}{2 - S(R + 1 + \sqrt{R^2 + 1} - 2M\sqrt{R^2 + 1}) \exp - m_2A'}$$
(29)

Equation (13) is rewritten as

$$m_2 A' = \frac{U'A'}{wc} \left(\frac{\sqrt{R^2 + 1} - R}{2} \right)$$
 (30)

Equation (29) requires a trial-and-error solution, which can best be accomplished by letting $\exp{-m_2A'}=1$ for the first trial. Then when a value of U'A'/wc becomes available from (29) it can be inserted into (30) and correction made if necessary. Since R is usually quite a large number, (30) becomes very small and it is rarely necessary to make a second trial with Equation (29); for example if R is 10 and U'A'/wc is 0.1 then $\exp{-m_2A'}$ is 0.9975.

It is worth noting that if there is no condensation M = 0 and (29) reduces to Underwood's (2) equation.

DETERMINATION OF INTERMEDIATE TEMPERATURES

In order to analyze the condensation section it becomes necessary to find t_1' and t_2' .

Rewrite (5) and equate this to (11):

$$\frac{2wc}{U'}\frac{dt^{I}}{dA} + t^{I} = T_2 - \frac{W\lambda}{WC} + G_1 \exp m_1 A + G_2 \exp m_2 A \quad (31)$$

Evaluate (32) at the middle where A = A', $t^{\text{I}} = t_1'$; also substitute (21) and (22) and put in dimensionless form with the aid of (17) and (25).

$$\frac{T_1 - t_1'}{t_2 - t_1} = \left(\frac{R}{S}\right) \frac{2 - S(R + 1 - \sqrt{R^2 + 1})}{2\sqrt{R^2 + 1} (R - 1 + \sqrt{R^2 + 1})} \left[\exp m_1 A' - \exp\left(-\frac{U'A'}{2wc}\right)\right] \\
- \left(\frac{R}{S}\right) \frac{2 - S(R + 1 + \sqrt{R^2 + 1})}{2\sqrt{R^2 + 1} (R - 1 - \sqrt{R^2 + 1})} \left[\exp m_2 A' - \exp\left(-\frac{U'A'}{2wc}\right)\right] \\
+ \left(\frac{1}{S} - R\right) \exp\left(-\frac{U'A'}{2wc}\right) + R$$
(33)

This differential equation (31) may be solved with the aid of the integrating factor $\exp(U'A/2wc)$ to yield the solution

$$T_{2} - t^{1} = \frac{G_{1}}{R - 1 + \sqrt{R^{2} + 1}}$$

$$\left[\exp m_{1}A - \exp\left(-\frac{U'A}{2wc}\right)\right]$$

$$+ \frac{G_{2}}{R - 1 - \sqrt{R^{2} + 1}}$$

$$\left[\exp m_{2}A - \exp\left(-\frac{U'A}{2wc}\right)\right]$$

$$+ \frac{W\lambda}{WC}\left[1 - \exp\left(-\frac{U'A}{2wc}\right)\right]$$

$$+ (T_{2} - t_{1}) \exp\left(-\frac{U'A}{2wc}\right)$$
 (32)

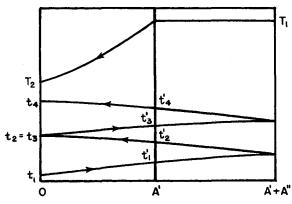


Fig. 6. Condenser subcooler.

Equation (12) is rewritten as

$$m_1 A' = -\frac{U'A'}{wc} \left(\frac{\sqrt{R^2 + 1} + R}{2} \right) (34)$$

 m_2A' is given by (30), and (33) gives the value of t_1' . In order to find t_2' rewrite (2).

$$t_2' - t_1' = \frac{W\lambda}{wc} \tag{35}$$

The logarithmic mean formula applies to the condensing section on the right side of Figure 1.

$$\frac{U''A''}{wc} = \ln\left(\frac{T_2 - t_1'}{T_2 - t_2'}\right) \quad (36)$$

The unknown in most practical problems is either U or A. Equations (29) and (36) give the dimensionless ratios UA/wc for their respective sections of the exchanger. The U's or the A's may be calculated directly from these ratios. If one wishes to calculate the true mean driving force for purposes of comparison, the over-all average rate expression may be set up and rearranged:

$$\Delta t = \frac{t_2 - t_1}{\left(\frac{UA}{wc}\right)} \tag{37}$$

In a later section it will be desirable to have the temperature t_r at the tube return-bend header. Setting up logarithmic mean formulas for both passes and equating the two yields

$$T_2 - t_r = \sqrt{(T_2 - t_1')(T_2 - t_2')}$$
 (38)

OTHER ARRANGEMENTS OF 1-2-PASS EXCHANGERS

Figure 2 shows a condenser subcooler. If the derivation is repeated in an analogous manner, the pertinent equations are

$$\frac{U'A'}{wc} = \frac{1}{\sqrt{R^2 + 1}} \ln \frac{2 - S(R + 1 - \sqrt{R^2 + 1} - 2RM + 2M\sqrt{R^2 + 1} \exp - m_2 A')}{2 - S(R + 1 + \sqrt{R^2 + 1} - 2RM)}$$

$$\frac{T_1 - t_1'}{t_2 - t_1} = -\left(\frac{R}{S}\right) \frac{2 - S(R + 1 + \sqrt{R^2 + 1} - 2RM)}{2\sqrt{R^2 + 1}(R + 1 + \sqrt{R^2 + 1})} \left[\exp m_1 A' - \exp\left(-\frac{U'A'}{2wc}\right)\right]$$

$$+\left(\frac{R}{S}\right) \frac{2 - S(R + 1 - \sqrt{R^2 + 1} - 2RM)}{2\sqrt{R^2 + 1}(R + 1 - \sqrt{R^2 + 1})} \left[\exp m_2 A' - \exp\left(-\frac{U'A'}{2wc}\right)\right]$$

$$+\left(\frac{1}{S} + RM\right) \exp\left(-\frac{U'A'}{2wc}\right) - RM$$
(40)

$$\frac{U''A''}{wc} = \ln\left(\frac{T_1 - t_1'}{T_1 - t_2'}\right)$$
 (41)

where

$$m_1 A' = \frac{U'A'}{wc} \left(\frac{\sqrt{R^2 + 1} + R}{2} \right)$$
 (42)

$$m_2 A' = -\frac{U'A'}{wc} \left(\frac{\sqrt{R^2 + 1} - R}{2} \right) (43)$$

$$T_1 - t_r = \sqrt{(T_1 - t_1')(T_1 - t_2')}$$
 (44)

 t_2 ' is given by Equation (35) and the other symbols are the same as before. In these equations the RM terms arise because the shell-side fluid and the tubeside fluid enter at opposite ends of the exchanger.

The heater-boiler arrangement shown in Figure 3 is solved with the same equations as the desuperheater condenser of Figure 1 if the following symbols are interchanged: w and W, c and C, t_1 and T_1 , t_2 and T_2 , t_1' and T_1' , t_2' and T_2' and the sign of λ is changed. The temperature-distribution diagram is inverted from Figure 1 to Figure 3.

The boiler-superheater arrangement shown in Figure 4 is solved with the same equations as is the condenser-subcooler arrangement of Figure 2 by interchanging the symbols as above.

EQUATIONS FOR USE WITH 1-4-PASS EXCHANGERS

Equations* for use with 1-4-pass exchangers have been derived because it is sometimes necessary to increase the tube-side fluid velocity when this becomes the limiting factor in design. The procedure is somewhat more complicated than for 1-2-pass exchangers.

Figure 5 shows the temperature distribution in a desuperheater condenser analogous to that of Figure 1. Any of the diagrams of Figures 5, 6, 7, or 8 may be inverted and the equations apply when the following symbols are interchanged: w and W, c and C, t_1 and T_1 , t_2 and T_2 , t_3 and T_3 , t_4 and T_4 , t_1' and T_1' , t_2' and T_2' , t_3' and T_3' , t_4' and T_4' , and the sign of λ is changed.

1-2- AND 1-4-PASS EXCHANGERS COMPARED

Comparison of the area requirements of 1-2- and 1-4-pass-exchanger arrangements has been made in two practical cases, a steam condenser subcooler with cooling water in the tubes and an ether desuperheater condenser using cooling water in the tubes. If the two over-all

coefficients of heat transfer U' and U'' were equal, the area required would be the same for 1-2- or 1-4-pass exchangers, and the shell-side-nozzle orientation would not make any difference. In view of the fact that the over-all coefficient of heat transfer for the cooling section is likely to be lower than that for the condensing section, the area requirements of the cooling section become of primary

importance. In both problems investigated the same order of preference prevailed.

For the ether desuperheater condenser the 1-2-pass exchanger of Figure 1 required the least area for desuperheating followed in order by the 1-4-pass exchanger of Figure 5, the 1-4-pass exchanger of Figure 7, and the 1-2-pass exchanger analyzed by Kern (1). The steam con-

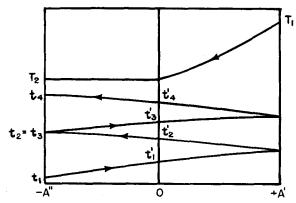


Fig. 7. Desuperheater condenser.

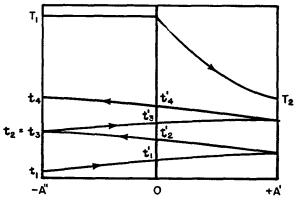


Fig. 8. Condenser subcooler.

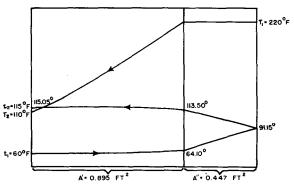


Fig. 9. Numerical example.

^{*}These equations, numbered (45) through (83), have been filed as document 4796 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for \$1.25 for photoprints or 35-mm. microfilm.

TABLE 1. COMPARISON OF HEAT EXCHANGER ARRANGEMENTS

Figure	Number of passes	Cooling section,	Condensing section,	Total,
	P	U'A'/wc	U''A''/wc	U'A'/wc + U''A''/wc
	Ethyl eth	er desuperheater	condenser	
1	1-2	0.0842	1.234	1.3182
5	1-4	0.0896	1.221	1.3106
7	1-4	0.0936	1.218	1.3116
Kern	1–2	0.0981	1.214	1.3121
	Stea	m condenser sub-	cooler	
2	1-2	0.0905	0.3837	0.4742
6	1-4	0.0927	0.3818	0.4745
8	1-4	0.0931	0.3818	0.4749
Kern	1-2	0.0967	0.3778	0.4745

denser subcooler required the least area for subcooling for the 1-2-pass exchanger of Figure 2 followed in order by the 1-4-pass exchanger of Figure 6, the 1-4-pass exchanger of Figure 8, and the 1-2-pass exchanger analyzed by Kern (1). Probably 1-6-pass and higher exchangers would lie between the two 1-4-pass exchanger arrangements.

Table 1 shows the results of the calculations. The differences between the various arrangements are only about 9% in the extreme cases, and so other considerations such as ease of piping or standardization of heat exchangers may be of greater importance.

In working with these equations it is necessary to have a consistent set of data to satisfy the assumption of negligible heat losses. In design work this requirement can be met. When exchangers are tested experimentally, there is usually some heat loss. However, if the heat balance be altered in such a way that there is a minimum of change in the temperatures, the results will be satisfactory.

A NUMERICAL EXAMPLE

Examination of Figures 2, 4, and 6 shows that the exit temperatures can cross. This is an undesirable condition because heat transfer surface is wasted. However, if a condenser operates temporarily at low load and the exit temperatures do cross, the equations are still applicable, as this example shows.

A vertical 1-2-pass heat exchanger condensing saturated steam at 220°F. and cooling the condensate to 110°F. may be considered. Steam flow amounts to 30 lb./hr. Cooling water passes through the tube side at the rate of 586 lb./hr., entering at 60° and leaving at 115°F. This is a 5° cross of the outlet temperatures. Table 2 summarizes the calculation of pertinent data, and Figure 9 shows the distribution of temperatures.

Estimations of the over-all coefficients for the cooling section U' and for the condensing section U'' were made by the usual empirical methods.

If the change in shell-side temperature had been neglected and a logarithmic mean temperature driving force used which involved the saturation temperature of the steam and the entering and leaving coolingwater temperatures, the result would have been U'A' + U''A'' = 246.7 B.t.u./(hr.) (°F.), an error of 11%.

SUMMARY

Equations have been presented for determining the temperature distribution and the ratios UA/wc for both cooling and condensing or heating and boiling sections of 1-2- and 1-4-pass heat exchangers. Systematic procedures for using these equations have also been worked out. Comparison of two practical designs showed that the vertical 1-2-pass exchanger with cooling section next to the channel end requires the least area, followed in decreasing order of preference by the 1-4-pass exchanger with cooling next to the channel end, the 1-4-pass

exchanger with condensing next to the channel end, and the 1-2-pass exchanger with condensing next to the channel end. It may be presumed that 1-6- and higher pass exchangers lie between the 1-4-pass arrangements.

ACKNOWLEDGMENT

Grateful acknowledgment is due the Nebraska Engineering Experiment Station for funds to support that portion of the work involving 1-4-pass exchangers.

NOTATION

Capital letters refer to the hot fluid, lower case letters to the cold fluid. Subscript 1 refers to the inlet, subscript 2 to the outlet.

4 = exposed tube area, sq. ft.

A' = tube area exposed in the temperature-change section of the exchanger, sq. ft.

A" = tube area exposed in the phasechange section of the exchanger, sq. ft.

c, C = fluid heat capacity, B.t.u./(lb.)
(°F.)

d = differential exp = exponential

g, G = integration constants

λ = latent heat of vaporization, B.t.u./lb.

ln = natural logarithm

m = roots defined by Equations (30), (34), (42), (43), (54), (55), (62), (63), (71), (72), (81), and (82)

M = latent heat load/total heat load of the exchanger

Q = parameter defined by Equation (46)

R = wc/WC

S = parameter defined by Equation (25) or (45)

 t^{I} , t^{II} , etc. = cold-fluid temperature in the tube passes I, II, etc., °F.

t₁', t₂', etc. = cold-fluid temperatures in the tube passes at the boundary between the sections, °F.

t_r = cold-fluid temperature at the tube return header, °F.

 Δt = true-mean-temperature-difference driving force, °F.

T = hot-fluid temperature, °F.

U' = over-all heat transfer coefficient for the cooling or heating section of the exchanger, B.t.u./(hr.) (sq. ft.)(°F.)

U" = over-all heat transfer coefficient for the condensing or boiling section of the exchanger, B.t.u./ (hr.)(sq. ft.)(°F.)

w, W = rate of fluid flow, lb./hr.

TABLE 2

$M = 0.898 \, [\text{Eq. (26)}]$ $m_2 \Lambda' = -0.00115 \, [\text{Eq. (43)}]$ $\exp -m^2 \Lambda' = 1.0011$ $m_1 \Lambda' = 1.791 \, [\text{Eq. (42)}]$

 $C = 1 \text{ B.t.u.}/(\text{lb.})(^{\circ}\text{F.})$

W = 30 lb./hr.

 $\lambda = 965 \text{ B.t.u./lb.}$

 $\exp m_1 A' = 5.993$

U'' = 500 B.t.u./(hr.)(sq. ft.)(°F.)A'' = 0.447 sq. ft.

LITERATURE CITED

- Kern, D. Q., "Process Heat Transfer," p. 283, McGraw-Hill Book Company, Inc., New York (1950).
- 2. Underwood, A. J. V., J. Inst. Petroleum Tech., 20, 145 (1934).