Concentration and Temperature Profiles in a Tubular Reactor for Methanol Synthesis with the Outer-wall of a Uniform Temperature. II. Profiles in Reactors with a Heat Exchanger Tube in the Catalyst Bed

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For better performance in methanol synthesis of the previously described tubular reactor, which was placed in a heat transfer medium of flownig oil and which was packed with a catalyst bed with no heat exchanger tube, it was pointed out that any large magnitude of temperature rise along the catalyst bed should be reduced[1]. The use of a heat exchanger tube in the catalyst bed was naturally expected to be a means of providing suitable control of the bed temperature. In this connection, several workers, such as Kodama et al.[2,3], Shindo et al.[4] and Matsuyama et al.[5], have discussed the temperature profiles in catalytic reactors equipped with various types of heat exchanger tubes, but their papers have provided no detailed experimental data on this subject. On the other hand, Kjaer[6] provided the data on the basis of which he had calculated the temperature profiles in both the catalyst bed and the heat exchanger tube, with an industrial reactor for the ammonia synthesis, but his treatment did not deal with the comparative merits of the different types of heat exchanger tubes.

In this paper, the experimental data on the profiles of methanol concentration and on the temperature measured with reactors which were individually equipped with three different types of heat exchanger tubes are presented, and their effects upon the profiles, as well as on the methanol yield, are discussed.

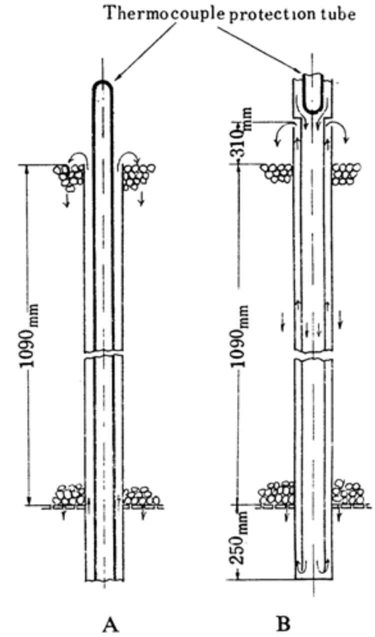

Fig. 1A. Heat exchanger tube (Type A and Type B).

Experimental

The same methanol synthesis reactor as was described in the preceding paper[1] was equipped successively with three different types of heat exchanger tubes, all three types of heat exchanger tubes were copper; the first and second ones were countercurrent types (types A and B), while the last one was a concurrent type (type C). Detailed sketches of them are shown in Figs. 1A and 1B. The A-type heat exchanger tube was a single tube of 10 mm. O. D. and 8 mm. I. D.; it was placed at the center-axis of the reactor. A thermocouple protection tube of stainless steel, 5 mm. O. D. with a thickness of 1 mm., went coaxially through the heat exchanger tube and permitted scanning of the temperature along the whole length by moving the thermocouple. The heat exchangers of types B and C were double tubes of the same dimensions. They consisted of an outer tube, sealed at one end, of 10 mm. O. D. and an inner tube of 6 mm. O. D., each of them having a thickness of 1 mm. For both the B and C types of heat exchangers, the thermocouple protection

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1) H. Uchida, Y. Ogino, M. Oba and K. Shimomura, This Bulletin, 35, 1400 (1962).

2) S. Kodama, K. Fukui and K. Takagi, *J. Chem. Soc. Japan, Ind. Chem. Sec.* (*Kōgyō Kagaku Zasshi*), 52, 123, 221 (1949).

3) S. Kodama, K. Fukui, K. Tame and K. Takagi, ibid., 53, 148 (1950).

4) M. Shindo and S. Araki, *Bull. Faculty Eng., Hokkaido Univ.*, 5, 143 (1951).

5) T. Matsuyama, Y. Sugawara and K. Matsuo, *Chem. Eng.* (*Kagaku Kikai*). 14, 229 (1950).

6) J. Kjaer, Measurement and Calculation of Temperature and Conversion in Fixed-bed Catalytic Reactors," Jul. Gjellerups Forlag, Copenhagen (1958).

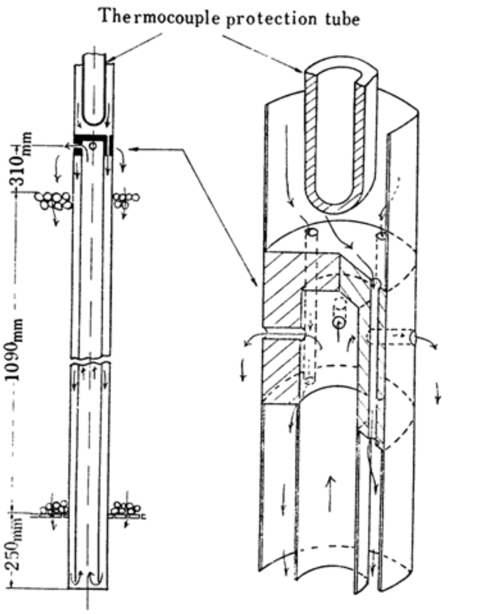

Fig. 1B. Heat exchanger tube (Type C).

tube was too thick to be inserted in the inner tube. The path of the gas stream in the reactor is indicated by arrows in the figures. It should be noted that, in the B-type heat exchanger, the synthesis gas introduced into the reactor at the top first flows downwards through the inner tube, whereas in the C-type exchanger it first flows downwards through the annular space between the outer and the inner tubes. For the sake of simplicity, the previous reactor with no heat exchanger will be hereafter denoted as the O reactor, while the individual reactors equipped with the heat exchangers of type A, type B, and type C will be termed the A, B, and C reactors respectively.

The catalyst used in the present study was a three-component catalyst (ZnO-Cr_2O_3-CuO) of the same composition and prepared by the same procedure as those of the catalyst in the preceding study[1]. Two liters of the reduced catalyst granules (3 mm. in diameter by 4 mm. high), weighing 3.012 kg., were charged into the reactor, and the catalyst bed attained as high as 1090 mm., occupying the intermediate range of the reactor between 1440 and 350 mm. from the bottom.

The operating procedure of the methanol synthesis was quite the same as has been described previously[1]. In this study, however, the bed temperature profile along the center-axis could not be measured, because the heat exchanger tube occupied the center-axis. As it was, only the off-center profiles at radial distances of 15 and 22 mm. from the center-axis were measured by means of two thermocouples in the protection tubes (5 mm. O. D. with a 1 mm. thickness) placed parallel in the bed.

The axial concentration profile was determined in the same way as in the preceding study[1], but the catalyst bed heights at which small portions of the fluid were withdrawn for the composition determinations varied somewhat according to the kinds of reactors[7].

Results

Methanol Production.—The methanol yields are listed in Table I, together with the operation variables. For the sake of comparison, the table also includes the previous data on the synthesis runs (Exps. I and I') using the O reactor. The A reactor produced a much larger amount of methanol than the O reactor, and the space-time-yield attained as much as twice the yield of the O reactor (compare Exp. I with Exp. 3), provided the synthesis gas was preheated at a higher temperature (cf. the data of Exps. 2 and 3). The B reactor could also produce a larger amount of methanol than the O reactor, but the production increase was rather low compared with that of the A reactor (compare Exp. 1 with Exp. 6). Contrary to the above findings, reliable performance data could not be collected with the C reactor until the introducing gas had been preheated to a much higher temperature (cf. Exp. 7). In this reactor, the heat exchanger tube cooled the catalyst bed so much that the bed temperature would decrease too low to allow the progress of the synthesis.

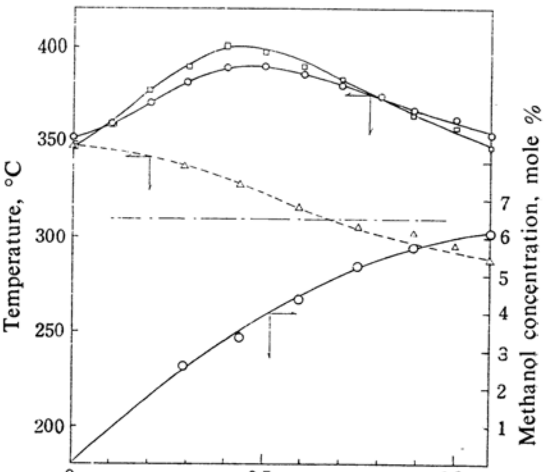

Fig. 2. Axial temperature and concentration profiles. Exp. 4.
—□— Off-center temperature profile at $r=15$ mm.
—○— Off-center temperature profile at $r=22$ mm.
…△… Temperature profile in heat exchanger tube.
—·—· Oil bath temperature.
—○— Methanol concentration profile.

7) The bed heights will be seen by the lengths of the abscissa corresponding to the marks along the experimental concentration profiles.

TABLE I. METHANOL YIELD AND OPERATION VARIABLES

Exp.	Heat exchanger	Flow rate of inlet gas l./hr.	Composition of inlet gas H_2/CO	Oil bath temp. °C	Preheated temp.[b] °C	Methanol yield kg./day	Space-time-yield of reactor kg./l. hr.
1	Type A	32170	2.92 (11.0)[a]	310	210	89	1.86
2	Type A	27810	2.92 (9.4)	315	208	90	1.87
3	Type A	56650	2.71 (13.3)	310	275	104.5	2.27
4	Type A	54570	4.59 (10.2)	310	275	101	2.11
5	Type B	32080	4.40 (3.3)	310	240	69	1.44
6	Type B	32170	3.15 (7.2)	302	222	67.5	1.41
7	Type C	44050	2.50 (9.2)	310	310	80.5	1.68
I	none	34470	3.0 (7.7)	295	295	42.2[c]	1.01
I′	none	33570	8.5 (10.5)	295	219	50.6[c]	1.21

Every run was made at 200 kg./cm^2 of the synthesis gas.
a) Numerals in the parentheses give the inert gas content (vol. %).
b) The temperature was measured immediately after the preheater.
c) In Exps. I and I′, 1.74 l. of the reduced catalyst was used.

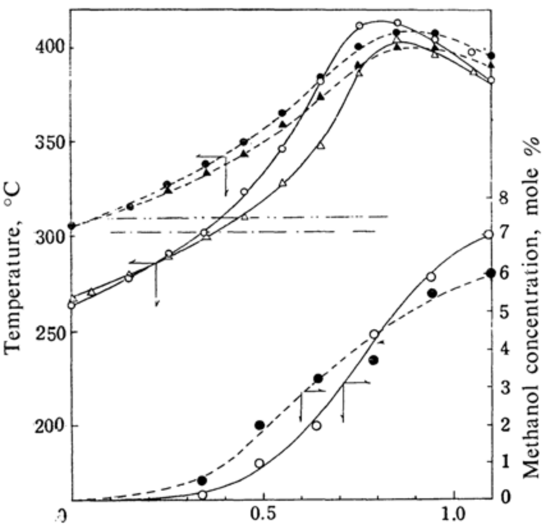

Fig. 3. Axial temperature and concentration profiles. Exps. 6 and 7.

Off-center temperature profile at r=15 mm.	—○—	Exp. 6.
	···●···	Exp. 7.
Off-center temperature profile at r=22 mm.	—△—	Exp. 6.
	···▲···	Exp. 7.
Oil bath temperature.	—·—	Exp. 6.
	—··—	Exp. 7.

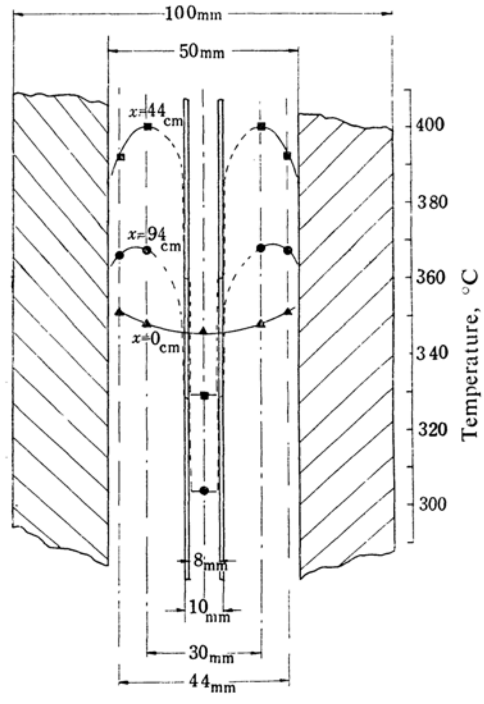

Fig. 4. Radial temperature profile.

Concentration and Temperature Profiles.—Figure 2 shows the concentration and temperature profiles for the synthesis run with the A reactor (Exp. 4), while Fig. 3 shows the profiles for the runs with the B reactor (Exp. 6) and the C reactor (Exp. 7.). The major difference observed among the temperature profiles of the three reactors is that the maximum of the temperature profile appears in the upper half of the catalyst bed of A reactor whereas it appears near the bottom of the bed of B and C reactors. Moreover, the temperature at the catalyst inlet of the B and C reactors is kept considerably lower than that of the A reactor even when the gas is preheated at higher temperatures. In the bed of the A reactor, methanol has already been produced at the top zone of the bed and increases in amount rather monotonously with the bed length, whereas in the beds of reactors B and C only a small amount of methanol begins to appear after the gas has passed through more than a quarter of the bed length. The concentration and temperature profiles observed with the B and C reactors are essentially the same in tendency as those observed with the O reactor[1)].

The off-center temperatures, as well as the temperature in the heat exchanger tube measured at the different bed lengths of 0, 0.44, and 0.94 m., are given as full marks in Fig. 4 for Exp. 4. A rough estimation of the radial profile from the available data leads us to expect a steep temperature rise in the middle across the annular-shaped cylindrical bed.

Discussion

Heat Transfer between Gases in the Catalyst Bed and the Heat Exchanger Tube.—Since the synthesis runs using the A reactor provide the experimental axial temperature profiles in the heat exchanger tube as well as in the catalyst bed, a quantitative treatment of the heat transfer phenomena between the gas in the catalyst bed and the gas in the heat exchanger tube will be made with this reactor. The heat balance equation for the bulk catalyst has the form:

$$S_1C_{p1}G_1dt_1/dx=S_1v\Delta H-F_{12}U_{12}(t_1-t_2)-F_{1b}U_{1b}(t_1-t_b) \tag{1}$$

while the equation for the gas in the heat exchanger tube is given by;

$$S_2C_{p2}G_2dt_2/dx=F_{12}U_{12}(t_1-t_2) \tag{2}$$

In the equations, S is the cross sectional area, C_p is the heat capacity of the gas, G is the mass velocity of the gas, v is the reaction rate based on the unit volume of the catalyst bed, ΔH is the reaction heat, F is the cooling area per unit length, and U is the overall heat transfer coefficient. Subscripts 1, 2, and b indicate the catalyst bed, the heat exchanger tube, and the oil bath respectively.

Because of the complicated nature of the first term on the right hand-side of Eq. 1, it is necessary to use a step by step integration of the differential equations. Under the assumptions that $S_1C_{p1}G_1$ and $S_2C_{p2}G_2$ are identical and that v is constant within the step length Δx, the differential equations can be directly transformed to the following difference equations:

$$A(t_{1no}-t_{1ni})/\Delta x=\Delta Q_n-F_{12}U_{12}\{(t_{1no}+t_{1ni})-(t_{2no}+t_{2ni})\}/2-F_{1b}U_{1b}\{(t_{1no}+t_{1ni})/2-t_b\} \tag{3}$$

$$-A(t_{2no}-t_{2ni})/\Delta x=F_{12}U_{12}\{(t_{1no}+t_{1ni})-(t_{2no}+t_{2ni})\}/2 \tag{4}$$

In Eqs. 3 and 4, A stands for $S_1C_{p1}G_1=S_2C_{p2}G_2$, ΔQ_n for $S_1v_n\Delta H$ where v_n is the v in the n-th step, and the subscripts ni and no indicate the inlet and outlet of the n-th step respectively. The difference equations are solved to result in the following respective expressions of $U_{12}F_{12}/A$, t_{1no}, and t_{2no}:

$$U_{12}F_{12}/A=(t_{2ni}-t_{2no})/\Delta x\{(t_{1ni}-t_{2ni})+\Delta t/2\} \tag{5}$$

$$t_{1no}=b/a+t_{2no}c/a \tag{6}$$

$$t_{2no}=(ap-lb)/(lc+aq) \tag{7}$$

in which the following abbreviations are introduced:

$$\Delta t=(t_{1no}-t_{1ni})-(t_{2no}-t_{2ni}),\ a=A+F_{1b}\cdot U_{1b}\Delta\chi/2,\ b=\Delta Q+A(t_{1ni}-t_{2ni})+F_{1b}U_{1b}\cdot\Delta x(t_b-t_{1ni}/2),\ l=B\Delta x/2,\ p=t_{2ni}\ (1\text{-}B\Delta x/2)-B\Delta x(t_{1ni}/2-t_{2ni}),\ q=(1-B\Delta x/2),$$
$$\Delta Q=\Delta Q_n\Delta x$$

The left-hand side term of Eq. 5 corresponds to the cooling coefficient of the heat exchanger tube and will be denoted by B hereafter.

Let us calculate B by the aid of Eq. 5 on the basis of the experimental profiles of Exps. 1, 2, 3, and 4. In the calculation, t_1 is conventionally chosen to fit the arithmetic mean temperature between the two measured off-center temperatures. The step lengths used in the calculation are 0.29 m. for the first step and 0.15 m. for the other steps, corresponding to the bed heights at which small portions of the fluid in the bed were drawn for the composition determination. The B thus obtained is denoted by B_{exp} and is shown in Fig. 6 as a function of x. The figure shows that B_{exp} is different according to the bed lengths and that a maximum appears at about $x=0.4$ m.

The B_{exp}'s are then averaged over the whole bed length (denoted by $\overline{B}_{\mathrm{exp}}$ hereafter), and the $\overline{B}_{\mathrm{exp}}$ is compared with the value of B_{calc}, which can be calculated by usual means. In the comparison, B_{calc} averaged over the whole bed length, for the sake of convenience, is taken into account[8]; it is usually given by the following formulae:

$$B_{\mathrm{calcd}}=U_{12}F_{12}/A \tag{8}$$

where U_{12} is defined either by

$$1/U_{12}=1/h+(1/h_a)(r_2/r_1)+r_2/\lambda_s\ln(r_2/r_1) \tag{9}$$

or by

$$1/U_{12}=1/h'+(1/h_a)(r_2/r_1)+r_2/\lambda_s\ln(r_2/r_1)+r_2/\lambda_e\ln(r_3/r_2) \tag{10}$$

In the above formulae, h is the heat transfer coefficient based upon the difference between the average catalyst temperature and the temperature of the outer-wall of the heat exchanger tube, whereas h' is the coefficient across

8) The calculation of individual B's at different bed lengths is very time-consuming, since the thermodynamic data must be evaluated at different bed lengths.

TABLE II. FORMULAE FOR EVALUATION OF h AND h'

Symbol	Formula	Ref.
A	$hD/k=21.3(R_{ep})^{0.6}\exp(-5.72d/D)d^{0.15}$	9)
B	$hD/k=4.9(R_{ep})^{0.6}\exp(-2.2d/D)$	10)
C	$hD/k=3.5(R_{ep})^{0.7}\exp(-4.6d/D)$	11)
D	$hD/k=0.813(R_{ep})^{0.9}\exp(-6d/D)$	12)
E	$h=60+0.083R_{ep}$	13)
F	$h'D/\lambda_e=254(R_{ep}d/D)^{-0.87}$ $\lambda_e/k=0.209(R_{ep})^{0.87}$	14)

TABLE III. THERMODYNAMIC DATA AND OTHER MATTERS

Exp.	C_p kcal./kg. °C	μ kg./m. hr.	k kcal./m. hr. °C	λ_e kcal./m. hr. °C	G kg./m². hr.	R_{ep} in catalyst bed	R_{ep} in heat exchanger tube
1	0.654	0.0972	0.204	6.15	8.41×10^3	3.03×10^2	3.62×10^3
2	0.701	0.0948	0.210	5.15	6.47×10^3	2.39×10^2	2.85×10^3
3	0.607	0.1001	0.205	10.47	15.90×10^3	5.56×10^2	6.64×10^3
4	0.800	0.0936	0.222	9.21	11.68×10^3	4.37×10^2	5.22×10^3

TABLE IV. HEAT TRANSFER COEFFICIENTS

Heat transfer coefficient kcal./m² hr. °C		Formula for evaluation	Exp. 1	Exp. 2	Exp. 3	Exp. 4
h_a	$\times10^{-2}$		6.36	6.07	16.54	15.57
h	$\times10^{-2}$	A	7.76	6.33	11.22	10.53
	$\times10^{-3}$	B	1.43	1.27	2.06	1.94
	$\times10^{-2}$	C	7.79	6.79	11.96	10.96
	$\times10^{-2}$	D	3.40	2.90	6.03	5.26
	$\times10$	E	8.51	7.98	10.62	9.63
h'	$\times10^{-3}$	F	2.70	2.78	2.71	2.94
U_{12}	$\times10^{-2}$	A	3.07	2.85	6.06	5.69
	$\times10^{-2}$	B	3.74	3.51	8.04	7.56
	$\times10^{-2}$	C	3.07	2.83	6.27	5.82
	$\times10^{-2}$	D	2.04	1.82	4.14	3.69
	$\times10$	E	7.29	6.85	9.82	9.22
	$\times10^{-2}$	F	2.97	2.74	5.76	5.45

the gas film on the outer surface of the tube, h_a is the coefficient from the cooling gas to the inner-wall of the tube, λ_s is the thermal conductivity of the heat exchanger tube material (315 kcal./m. hr. °C), λ_e is the effective thermal conductivity of the catalyst bed, r_1 and r_2 are the inner and the outer radii of the heat exchanger tube, and r_3 is the radius to give the area which is equivalent to a half of the cross sectional area of the catalyst bed. The last term on the right-hand side of Eq. 10 has no strict meaning in theory, but it is introduced conventionally because the $1/U_{12}$ should include any term of the heat transfer resistance through the catalyst bed. The term may stand for the mean value of the transfer resistance.

The evaluation of h can be made by the aid of several kinds of formulae (A[9], B[10], C[11], D[12], and E[13]), while h' can be evaluated by the aid of formula F[14], as listed in Table II. The symbols not explained earlier are as follows: R_{ep} is the modified Reynolds number, D is the equivalent diameter of the annular shaped bed, and μ is the viscosity of the gas. Moreover, either one of the following formulae can give h_a according as whether the gas flow is laminar or turbulent[15]:

9) S. Maeda and K. Kawazoe, *Chem. Eng.* (*Kagaku Kikai*), **15**, 5, 9, 312 (1951).
10) S. Hatta and S. Maeda, ibid., **12**, 56 (1948).
11) M. Leva, M. Weintaub, M. Gummer and E. L. Clark, *Ind. Eng. Chem.*, **40**, 747 (1948).
12) M. Leva, ibid., **39**, 857 (1947).
13) S. Sugawara and S. Sato, *Coll. Papers Chem. Eng.* (*Kagaku Kikai Ronbunshu*), 13, No. 45, 164 (1947).
14) S. Hatta and S. Maeda, *Chem. Eng.* (*Kagaku Kikai*), **12**, 56 (1948), **13**, 79 (1949).
15) In view of the magnitude of R_{ep} in Table III, the gas flow is laminar for the cases with Exps. 1 and 2, whereas it is turbulent for the cases with Exps. 3 and 4.

TABLE V. COMPARISON OF B_{calcd} WITH $\bar{B}_{\mathrm{exp}}$

Exp.	$\bar{B}_{\mathrm{exp}}$ m^{-1}	B_{calc} from formula A	B	C	D	E	F
1	1.005	0.954	1.163	0.955	0.630	0.227	0.922
2	0.880	1.077	1.325	1.068	0.685	0.259	1.033
3	0.837	1.073	1.423	1.110	0.732	0.174	1.020
4	0.942	1.046	1.388	1.068	0.678	0.169	0.999

TABLE VI. HEAT TRANSFER RESISTANCES

Exp.	$1/U_{12}$	$1/h'$	$(1/h_a)(r_2/r_1)$	$(r_2/\lambda_s)\ln(r_2/r_1)$	$(r_2/\lambda_e)\ln(r_3/r_2)$
1	3.370×10^{-3}	3.701×10^{-4}	1.967×10^{-3}	3.537×10^{-6}	1.026×10^{-3}
4	1.837×10^{-3}	3.401×10^{-4}	8.029×10^{-4}	3.537×10^{-6}	6.867×10^{-4}

$$h_a = 1.75(k/D_e)(C_pG/SkL)^{1/3} \quad \text{for a laminar flow} \qquad (11)$$

$$h_a = 0.023(k/D_e)(D_eG/\mu)^{0.8}(C_p\mu/k)^{0.4} \quad \text{for a turbulent flow}^{16)} \qquad (12)$$

where k is the thermal conductivity of the gas, D_e is the equivalent diameter of the heat exchanger tube, and L is the catalyst bed length. Table III summarizes the thermodynamic and other data with respect to Exps. 1, 2, 3 and 4. In Table V, $\bar{B}_{\mathrm{exp}}$ is compared with the B_{calcd} obtained by putting into Eq. 8 the values of the heat transfer coefficients as listed in Table IV. A comparison shows that the B_{calcd}'s obtained by the use of h's from the A, C, and E formulae, and also the B_{calcd} obtained by the use of h' from formula F, are consistent with the $\bar{B}_{\mathrm{exp}}$.

In addition, the calculation of $1/U_{12}$ by means of Eq. 10 provides the content of the overall heat transfer resistance between the gas in the catalyst bed and the gas in the heat exchanger tube; this is shown in Table VI. The table shows that the heat transfer resistance across the catalyst bed is so high as to account for about one-third of the overall heat transfer resistance.

Calculation of Axial Temperature Profiles in the A Reactor.—Under the assumption that the catalyst inlet temperature is identical with the temperature at the end of the heat exchanger tube, a step by step calculation of the axial temperature profiles in both the catalyst bed and the heat exchanger tube is possible with the aid of Eqs. 6 and 7, if ΔQ_n, U_{1b}, and B, besides the thermodynamic data, are available. In this connection, ΔQ_n is given as a function

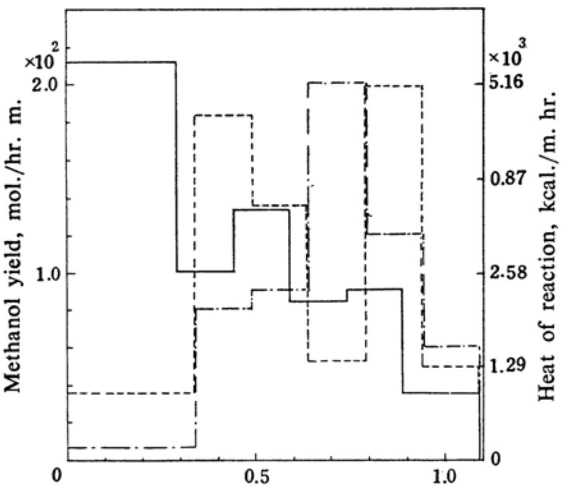

Fig. 5. Methanol yield and reaction heat of methanol synthesis as function of bed length.
—— Exp. 4. —·— Exp. 6. ······ Exp. 7.

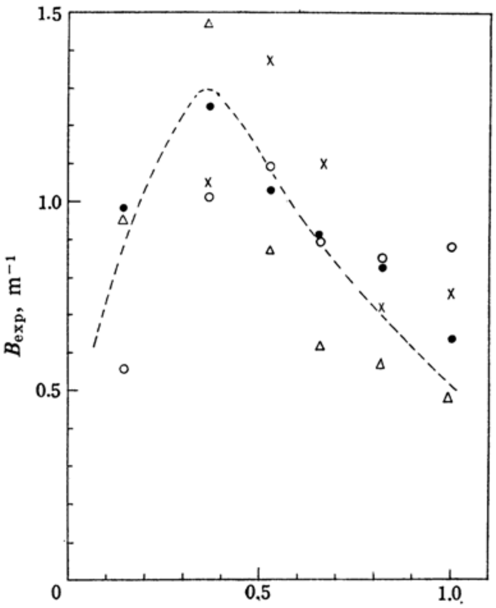

Fig. 6. Cooling coefficient as function of bed length.
× Exp. 1. ○ Exp. 2. △ Exp. 3. ● Exp. 4.

16) McAdams, "Heat Transmission", McGraw-Hill Book Co. Inc., New York (1954), pp. 219, 232.

of x in Fig. 5, while B_{exp} is given in Fig. 6 also as such. U_{1b} can be evaluated as a function of x by the following equation:

$$U_{1b}F_{1b}\{(t_{1ni}+t_{1no})/2-t_b\}\Delta x=\Delta Q-A\Delta t \tag{13}$$

For the sake of simplicity, however, B_{calcd} evaluated from the formula of Leva et al.[11] and U_{1b} averaged over the whole bed length are used in the present calculations. The choice of B_{calcd} is due to the afore-mentioned finding that the evaluated B_{calcd} was very approximate to the $\bar{B}_{exp.}$. The calculations are now made on the basis of the data of Exp. 4[17], and the resultant profiles are shown in Fig. 7 in comparison with the experimental ones. The accordance between the two sets of profiles is good. This indicates that an adequate value of B_{calcd} evaluated from any of the formulae thus far proposed can be used for the profile calculation without causing any serious errors.

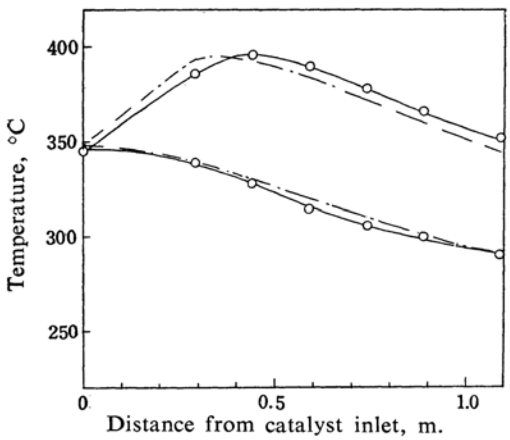

Fig. 7. Comparison between experimental and calculated temperature profiles. Exp. 4.
—○— Experimental profile.
—·—· Calculated profile.

Radial Temperature Profile across the Reactor. —Because only four measurements of the temperatures, namely, the temperatures at the center axis of the heat exchanger tube, the off-center temperatures at $r=15$ and 22 mm. in the catalyst bed, and the oil bath temperature, were made at several different bed lengths, the experimental radial profile across the reactor has not yet been obtained. In view of the estimated value of h', as well as of those of the respective heat transfer coefficients across the films on the outer and the inner surfaces of the reactor (h_L and h'')[18], steep temperature rises are expected at the outer surface of the heat exchanger tube and at the inner and the outer surface of the reactor. However, nothing decisive can be said in this respect.

Axial Temperature Profiles in B-Type and C-Type Heat Exchanger Tubes.—An attempt has been made to calculate the temperature profiles in the heat exchanger tubes of the B and C types. The calculations have been made by starting with a set of the following heat balance equations (14, 15, and 16):

$$Adt_1/dx=\Delta Q-F_{1b}U_{1b}(t_1-t_b)-F_{12}U_{12}(t_1-t_2) \tag{14}$$

$$\mp Adt_2/dx=F_{12}U_{12}(t_1-t_2)-F_{23}U_{23}(t_2-t_3) \tag{15}$$

$$\pm Adt_3/dx=F_{23}U_{23}(t_2-t_3) \tag{16}$$

The respective minus and plus signs on the left hand-side of Eqs. 15 and 16 are used as with the B-type heat exchanger tube, while the corresponding plus and minus signs are as with the C type one. The subscript 1 indicates the catalyst bed, as has been mentioned, and the subscripts 2 and 3 indicate the annular-shaped space and the inner tube of a double tube heat exchanger respectively. Since the thermodynamic as well as the mechanical data, such as C_p, U, G ΔQ, S and F, have already been

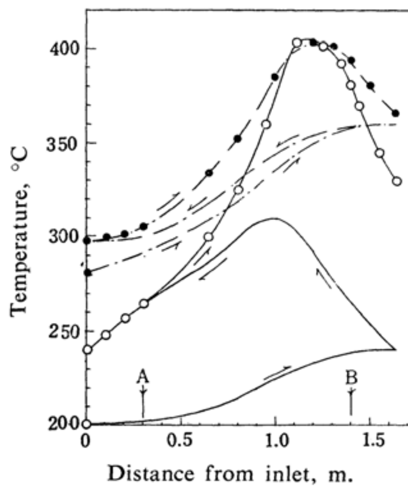

Fig. 8. Calculated temperature profiles in heat exchanger tube. Exps. 6 and 7.
Exp. 6. ——— Calculated profile in heat heat exchanger tube.
—○— Experimental profile in catalyst bed.
Exp. 7. —·—· Calculated profile in heat exchanger tube.
—·●—· Experimental profile in catalyst bed.
A; catalyst inlet, B; catalyst outlet.

17) In Exp. 4, the catalyst inlet temperature was 348°C. Evaluation of U_{1b} and B_{calcd} resulted in 1.524×10^2 kcal./m²hr.°C and 1.0684 m⁻¹ respectively.

18) The value of h_L was estimated to be 2.091×10^2 kcal./m² hr. °C (cf. 1)), while that of h'' is evaluated, for instance, to be 2.465×10^3 kcal./m² hr. °C for the case of Exp. 4.

TABLE VII. HEAT TRANSFER COEFFICIENTS WITH B AND C TYPES HEAT EXCHANGER TUBES

Exp.	Passage I R_{ep}	Passage I h kcal./m². hr. °C	Passage I Formula for evaluation	Passage II R_{ep}	Passage II h_a kcal./m². hr. °C	Passage II Formula for evaluation
6	2.97×10^2	7.698×10^2	A in Table II	3.55×10^3	6.361×10^2	(11) in this paper
7	4.05×10^2	9.072×10^2	A in Table II	4.84×10^3	1.302×10^3	(11) in this paper

Exp.	Passage III R_{ep}	Passage III h_a kcal./m² hr. °C	Passage III Formula for evaluation	Overall heat transfer coefficient U_{12} kcal./m² hr. °C	U_{23}	U_{1b}
6	1.24×10^4	1.399×10^3	(12) in this paper	3.060×10^2	3.771×10^2	8×10^2
7	8.46×10^3	2.038×10^3	(12) in this paper	4.841×10^2	6.615×10^2	1.27×10^2

available and since dt_1/dx can readily be obtained from the experimental temperature profiles, t_2 can be calculated by the aid of Eq. 14. Once t_2 is obtained as a function of x, t_3 can be calculated by the substitution of the value of dt_2/dx into Eq. 15.

The calculations have been conducted on the basis of the experimental data of Exps. 6 and 7; the results are shown in Fig. 8. The values of the heat transfer coefficient necessary for the calculations are listed in Table VII, in which the symbols I, II, and III stand for the passages as indicated in Fig. 9. The figure and the table disclose that, owing to the efficient heat transfer between the gases in the II and III passages, the temperature profiles in the second passage always slope down markedly in the upper zone, and that, as a consequence, the catalyst inlet temperature is kept comparatively low. These facts may have led to the axial temperature profile in the catalyst bed being almost identical in tendency with that for the reactor with no heat exchanger tube. Taking the most favorable temperature profile for the methanol synthesis (cf. 1)) into consideration, any remarkable effect in increasing the methanol yield is scarcely to be expected from the use of heat exchanger tubes of the B and C types, unless special treatment lowers the U_{23} in the upper part of the tubes. This may have resulted in the lower methanol yields of the B and C reactors as compared with the yield of the A reactor.

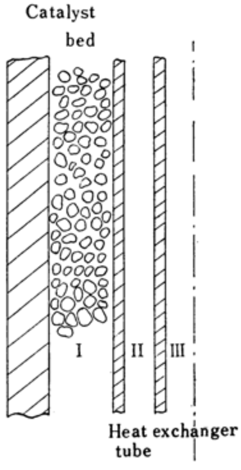

Fig. 9. Schematic diagram of flow passages.

Summary

Among the three types of heat exchanger tubes, a single tube countercurrent type, a double tube countercurrent type, and a double tube concurrent type, only the first one serves to increase the methanol production considerably compared with the effect of the previously tested reactor with no heat exchanger tube. On the basis of the experimental temperature profiles in both the catalyst bed and the single tube-heat exchanger tube, the cooling coefficient with the tube has been calculated to give the value as a function of the bed length. The value, when averaged over the whole bed length, agrees with the calculated value from the heat transfer coefficients, h or h' and h_a, available from certain formulae. By the use of this value, the average temperature profiles in the bed have been calculated on the basis of the experimental concentration profile; the resultant profile agrees well with the experimental one.

As regards the double tube-heat exchanger tubes, the temperature profiles for both the annular-shaped space and the inner tube have been calculated from the experimental bed temperature profiles. The results show that the profiles in the second passage of the tubes always slope down markedly in the upper zone and that, as a consequence, the bed temperature profiles are apt to remain almost identical with that for the previously tested reactor with no heat exchanger tube.

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