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THE HEIGHT EQUIVALENT TO A THEORETICAL PLATE OF RETENTIONLESS RECTANGULAR TUBES

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SUMMARY

Two different approaches to the evaluation of the height equivalent to a theoretical plate (HETP) of uniform tubes are reviewed and shown to be equivalent, and the latter is applied to the calculation of the HETP of tubes with a rectangular cross-section.

The dynamic diffusion constant and, by implication, the height equivalent to a theoretical plate (HETP) of retentive rectangular tubes were treated in a former publication[1] without regard to the "end effect" in tubes of finite width. This effect could be eliminated by bending the tube around and closing it on itself, so as to have, in effect, an annular tube, but the realization of such a structure would constitute a mechanical impossibility. This effect could be eliminated also by terminating both ends of the rectangular cross-section of the tube with, *e.g.*, an enlarged circular portion with a diameter of approximately one and a half times the thickness of the tube section, as illustrated by Fig. 2, so as to have a fluid flow near the end equal, on an average, to the average flow in the entire tube; but here again this would imply a construction not realizable when the tubes are formed by placing between two flat plates a thin sheet of flat material suitably cut out so that the rectangular tubes are constituted by the spaces left open between the two plates.

The specific purpose of this report is the determination of the increase in HETP due to the end effect in rectangular tubes with a large aspect ratio. This can be done with two methods which are quite different in approach, while being substantially equivalent, as will be shown first. Both methods are readily applicable to straight tubes with circular, elliptical, annular or rectangular cross-sections in which the viscosity controlled fluid has a velocity component in the direction of the tube axis.

The essential points of the first method for the case of a retentionless tube (see refs. 2–7) are as follows. The concentration $f(x, y, z, t)$ of any injected sample is inserted in the diffusion equation

$$D \triangle^2 f = \frac{df}{dt} = \frac{\partial f}{\partial t} + v \cdot \frac{\partial f}{\partial x} \tag{1a}$$

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where $\Delta^2 f$ denotes the laplacian of f in the y–z plane, and the solution of eqn. 1a is constrained by the boundary condition at the tube wall expressed by equating to zero the vectorial product

$$d\vec{s} \times \text{grad}\, f = 0 \tag{1b}$$

where $d\vec{s}$ designates a vectorial element of the cross-section periphery.

Let v_0 designate the average velocity and introduce the change of variable:

$$x_1 = x - v_0 t \tag{2}$$

Eqn. 1a becomes:

$$D\, \Delta^2 f = \frac{\partial f}{\partial t} + (v - v_0)\cdot\frac{\partial f}{\partial x_1} \tag{1c}$$

Let further $\bar{f}(x, t)$ designate the average concentration in any section of the tube with the abscissa x_1, and define Δf by:

$$f = \bar{f} + \Delta f \tag{3}$$

The application to both sides of eqn. 1c of the operator $(1/S) \int dS$, where the integral extends over the tube cross-section, gives:

$$D\cdot\frac{d^2\bar{f}}{dx_1^2} = \frac{\partial\bar{f}}{\partial t} + \frac{1}{S}\int (v - v_0)\cdot\frac{\partial\Delta f}{\partial x_1}\cdot dS \tag{4}$$

When f as given by eqn. 3 is merely substituted in 1c we obtain:

$$D\left(\Delta^2\, \Delta f + \frac{\partial^2\bar{f}}{\partial x_1^2} + \frac{\partial^2\Delta f}{\partial x_1^2}\right) = \frac{\partial\bar{f}}{\partial t} + \frac{\partial\Delta f}{\partial t} + (v - v_0)\left(\frac{\partial\bar{f}}{\partial x_1} + \frac{\partial\Delta f}{\partial x_1}\right) \tag{5}$$

Subtracting eqn. 4 from 5 and making the assumption that, after a sufficiently large diffusion time, we have $\Delta f \ll \bar{f}$ so that terms in Δf can be neglected against similar terms in f, yields:

$$D\, \Delta^2\, \Delta f = (v - v_0)\, (\partial\bar{f}/\partial x_1) \tag{6}$$

Since $\overline{v - v_0} = 0$, eqn. 6 admits a solution satisfying the boundary condition 1b; this solution can be written, symbolically

$$\Delta f = \frac{1}{D}\cdot\Delta^{-2}\, (v - v_0)\cdot\frac{\partial\bar{f}}{\partial x_1} \tag{6a}$$

and substitution of eqn. 6a in 4 and re-arranging the terms gives:

$$\left[D - \frac{1}{DS}\int (v - v_0)\, \Delta^{-2}\, (v - v_0)\, dS\right]\cdot\frac{\partial^2\bar{f}}{\partial x_1^2} = \frac{\partial\bar{f}}{\partial t} \tag{7}$$

Eqn. 7 indicates that after a sufficient time the diffusion process will take place as if it were controlled by the dynamic diffusion constant:

$$D_1 = D - (1/DS) \int (v - v_0) \triangle^{-2} (v - v_0) \, \mathrm{d}S \tag{8}$$

The second method consists in making first the same coordinate change defined by eqn. 2 and in postulating that after a sufficiently long time the concentration of a sample injected at time $t = 0$ can be represented by the expression

$$f = \frac{1}{\sqrt{t}} \cdot \exp\left[- \frac{(x_1 - e)^2}{4 \, D_1 t} \right] \tag{9}$$

where e designates a function of y and z.

When eqn. 9 is substituted in the diffusion equation 1c three groups of terms are obtained, some with the coefficient $(x_1 - e)^2$; some with the coefficient $(x_1 - e)$; and a third group with neither. Satisfying eqn. 1c independently with each group, the first and third group give the relation

$$D_1 = D \, [1 + (\partial e/\partial y)^2 + (\partial e/\partial z)^2] \tag{10}$$

and the second

$$\triangle^2 e = -(1/D) \, (v - v_0) \tag{11}$$

wherefrom we can write, symbolically

$$e = -(1/D) \, \triangle^{-2} (v - v_0) \tag{12}$$

to which must be added the boundary conditions at the retentionless wall similar to eqn. 1b:

$$\mathrm{d}\vec{s} \times \mathrm{grad} \, e = 0 \tag{13}$$

To the postulation expressed by eqn. 9 we must add now the assumption that after a sufficiently large diffusion time we obtain D_1 with good approximation by averaging the right-hand side of eqn. 10 over the tube cross-section:

$$D_1 = D + \frac{D}{S} \int \left[\left(\frac{\partial e}{\partial y} \right)^2 + \left(\frac{\partial e}{\partial t} \right)^2 \right] \mathrm{d}S \tag{14}$$

The equivalence of the two methods can be demonstrated by showing that the respective second terms of the right-hand side of eqns. 8 and 14 are equivalent. The second term of eqn. 10 can be written, with eqn. 11

$$-(D/S) \int e \, \triangle^2 \, e \, \mathrm{d}S \tag{15}$$

and we must show that we have:

$$\iint \left[e \, \triangle^2 \, e + \left(\frac{\partial e}{\partial x}\right)^2 + \left(\frac{\partial e}{\partial z}\right)^2 \right] = 0 \tag{16}$$

To this effect we note first that the right-hand side of eqn. 13 can be multiplied by the scalar quantity e

$$d\vec{s} \times e \cdot \mathrm{grad}\, e = 0$$

which is equivalent to writing for the entire cross-section

$$\int \triangle^2 \, e^2 \, dS = 0 \tag{17}$$

which, when developed, gives the left-hand side of eqn. 16, Q.E.D.

We apply now the second method to the determination of the HETP of a tube with the rectangular cross-section $2A \times 2B$ and a large aspect ratio.

A first task consists in determining the fluid velocity as a function of the y, z coordinates of the tube section. This velocity is related to the pressure drop by the classical expression

$$\mu \, \triangle^2 \, v = dp/dx \tag{18}$$

with the boundary conditions at the tube walls:

$$v = 0 \quad \text{at} \quad y = \pm A \quad \text{and} \quad z = \pm B \tag{19}$$

We shall restrict our attention to the case of a large aspect ratio, *i.e.*, when:

$$A/B \gg 1 \tag{20}$$

We normalize eqns. 18 and 19 and write

$$\triangle^2 \, v = -1 \tag{18a}$$

with the boundary conditions

$$v = 0 \quad \text{at} \quad y = \pm a\pi/2 \quad \text{and} \quad z = \pm \pi/2 \tag{19a}$$

for the case:

$$a \gg 1 \tag{20a}$$

We start from the identity

$$-1 = -\frac{16}{\pi^2} \sum_{m=0}^{\infty} \frac{(-1)^m}{2m+1} \cdot \cos\,(2m+1)\frac{y}{a} \cdot \sum_{n=0}^{\infty} \frac{(-1)^n}{2n+1} \times \cos\,(2n+1)\,z \tag{21}$$

and a simple integration with respect to the separated variables y and z gives us:

$$v(y, z) = \frac{16}{\pi^2} \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \times \frac{(-1)^{m+n} \cos \frac{(2m+1)}{a} y \cos (2n+1) z}{(2m+1)(2n+1)\left[\left(\frac{2m+1}{a}\right)^2 + (2n+1)^2\right]} \tag{22}$$

which satisfies eqns. 18a and 19a.

Integration of v over the entire tube cross-section gives us the total flow:

$$F = \int_{-\pi/2a}^{\pi/2a} \mathrm{d}y \int_{-\pi/2}^{\pi/2} v(y, z)\, \mathrm{d}z = \frac{64a}{\pi^2} \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \times$$

$$\times \frac{1}{(2m+1)^2 (2n+1)^2 \left[\left(\frac{2m+1}{a}\right)^2 (2n+1)^2\right]} \tag{23}$$

When $a \gg 1$ a first approximation for F gives us

$$F_1 = \frac{64a}{\pi^2} \sum_{m=0}^{\infty} \frac{1}{(2m+1)^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^4} = \frac{64a}{\pi^2} \cdot \frac{\pi^2}{8} \cdot \frac{\pi^4}{96} = \frac{\pi^4 a}{12} \tag{24}$$

which is what we expect in first approximation from a cross-section $\pi a \times \pi$ for $a \gg 1$. with v normalized as per eqns. 18a and 19a.

However, we know that at the extremities of the rectangular cross-section there will be a somewhat stagnant layer and that, even if, with growing a, the importance of this layer as a fraction of the entire flow decreases with $1/a$, its retentiveness having to diffuse over a distance which increases with a, its net effect will not be negligible. Therefore, we should evaluate the right-hand side of eqn. 23 with a net error which decreases with a, in order to obtain a correct assessment of the stagnant end-layer effect.

Toward this purpose we evaluate eqn. 23 slice by slice for each n value, and write:

$$F_2 = \frac{64a}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \sum_{m=0}^{\infty} \frac{1}{(2m+1)^2 \left[\left(\frac{2m+1}{a}\right)^2 + (2n+1)^2\right]}$$

$$= \frac{64a}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \sum\nolimits_1 \tag{23a}$$

We then approximate the remnant of $\sum_1$ with an integral starting at some value m_0 of m

$$\sum_1 = \sum_{m=0}^{m_0-1} \frac{1}{(2m+1)^2\left[\left(\frac{2m+1}{a}\right)^2 + (2n+1)^2\right]} + \int_{m_0}^{\infty} \frac{dm}{(2m)^2\left[\left(\frac{2m}{a}\right)^2 + (2n+1)^2\right]}$$

$$= \sum_{m=0}^{m_0-1} \frac{1}{(2m+1)^2\left[\left(\frac{2m+1}{a}\right)^2 + (2n+1)^2\right]} + \frac{1}{(2n+1)^2} \times \int_{m_0}^{\infty} \left[\frac{1}{(2m)^2} - \frac{1}{(2m)^2 + a^2(2n+1)^2}\right] dm$$

$$= \sum_{m=0}^{m_0-1} \frac{1}{(2m+1)^2\left[\left(\frac{2m+1}{a}\right)^2 + (2n+1)^2\right]} + \frac{1}{2a(2n+1)^3} \times \left[\frac{(2n+1)\,a}{2m_0} - \arctan\frac{(2n+1)\,a}{2m_0}\right] \qquad (25)$$

and we note that this approximation is correct to within less than a fraction, $1/4m_0^3$, of the dominant member of the last bracket.

We are interested in knowing within a fraction $1/a$ the departure from the right-hand side of eqn. 24 of the right-hand side of eqn. 23a when the right-hand side of eqn. 25 is substituted for $\sum_1$ in eqn. 23a. Therefore, we should select values of m_0 which are small enough so that the departure of the first term for $\sum_1$ from its value for very large values of a, when multiplied by a, decrease with a; yet m_0 must be large enough so that the fractional error of less than $1/4m_0^3$ caused by the substitution of an integral for the summation decreases also with a. Both requirements will be met if we select for m_0 a value of the order of $a^{1/3}$. In this case, the part of the right-hand side of eqn. 25 which, when multiplied by a, remains stationary, is the second term of the bracket which, being negative as it should, represents the value of $F_1 - F_2$ for large a values and constitutes a measure of the stagnancy of the layers at both ends of the rectangular cross-section.

We have therefore, for large a

$$-\triangle\textstyle\sum_1 = \pi/4a(n+1)^3$$

and

$$F_1 - F_2 = \frac{64a}{\pi^2}\sum_{n=0}^{\infty}\frac{\pi}{4a(n+1)^5} = \frac{16}{\pi}\cdot S_5 \tag{26}$$

where:

$$S_5 = \sum_{n=0}^{\infty}\frac{1}{(1+n)^5} = 1.00452 \tag{26a}$$

Division of $F_1 - F_2$ by $\pi^4/12$ gives the extent of the stagnant layer at each end of the cross-section as a fraction, ε, of the half tube thickness:

$$\varepsilon = \frac{16}{\pi}\cdot S_5\cdot\frac{12}{\pi^4} = 0.63024 \tag{27}$$

We know that the contribution of the stream velocity to the dynamic diffusion constant within a flat tube and without reckoning the end effect is the thickness term in eqn. 40 of ref. 1 for $k = 0$

$$\triangle D_a = D_1 - D = \frac{2}{105}\cdot\frac{v_0^2 z_0^2}{D} \tag{28}$$

where z_0 designates the half tube thickness, and we calculate the effect of the stagnant end layer by postulating that we have a simple superposition of the effect expressed in eqn. 28 and the end effect. Toward this purpose we assume that the flow proceeds as a block at the average velocity, v_0, while the sample can diffuse freely with the stagnant layer, and compute the derivative of the e function from eqn. 13. Since $v = 0$ in the stagnant layer, we have $e' = 0$ at the wall and $e' = (v_0/D)\cdot 0.63024\cdot z_0$ at the stagnant layer–stream interface. From there on e' decreases steadily in the stream which proceeds at a velocity which exceeds the velocity average by the amount $0.63024/a$ so that at the other end e' has the value -0.63024 after which it increases in the stagnant layer until it has again the value zero at the end wall.

The mean square value of e is therefore

$$\frac{1}{3}\left(0.63024\cdot\frac{v_0 z_0}{D}\right)^2$$

and its contribution, $\Delta D_{b'}$, to the dynamic diffusion constant will be therefore, in accordance with eqn. 14

$$\Delta D_{b'} = 0.13240 \cdot v_0^2 z_0^2 / D \tag{29}$$

which, when added to the regular thickness term gives:

$$D_1 = D + 0.13240 \cdot v_0^2 z_0^2 / D \tag{30}$$

By comparison with eqn. 28 we see that the end effect contribution to dynamic diffusion is 6.9512 times larger than the thickness term. It means also that the optimal HETP will be $\sqrt{6.9512 + 1} = 2.820$ times larger than that calculated on the basis of the thickness term only. Figs. 1 and 2 illustrate the surprisingly large diffusion difference between the modified tube and the regular rectangular tube.

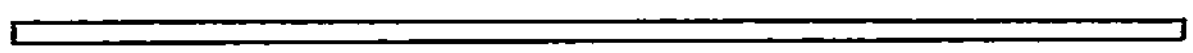

Fig. 1. Rectangular tube profile. $D_1 = D + 0.1514 \cdot v_0^2 z_0^2 / D$.

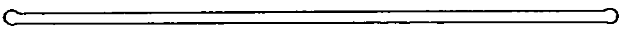

Fig. 2. Tube profile for minimization of end-effect. $D_1 \rightarrow D + 0.0190 \cdot v_0^2 z_0^2 / D$.

The highly critical dependency of the diffusion constant upon uniformity of the tube cross-section should be noted. Thus, assuming an aspect ratio of 100 for the tube illustrated by Fig. 2, if the two halves of the section were to differ by 1% in thickness, this would introduce another term in D_1 equal to 17.5 times the regular thickness term, and this effect would increase quadratically with the fractional difference between the two halves. This conclusion can be readily established by using formulae 12 and 14.

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