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MATHEMATICAL MODEL FOR MIXING OF FLUID IN PACKED BED

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1. Introduction

As fluid flows through packed bed, it suffers the split of streams around particles and the changes in velocity, and these behaviors cause the mixing of fluid. The mixing phenomenon is important because of its effect on the performance of packed-bed reactor and the efficiencies of mass transfer in packed bed.

Recently, increased attention has been focussed on the problem of the mixing in packed bed, and a number of investigations have been made to determine the mixing characteristics of fluid in packed bed.

In the case of diffusion model, mixing process is described by familiar type of differential equation analogous to Fick's diffusion equation, and the characteristic parameter of this model is a longitudinal dispersion coefficient. Many works on the longitudinal mixing have been conducted on the basis of diffusion model because of its simpleness in mathematical treatment. But it may be guessed that this model does not fully describe the actual behavior of fluid flowing through packed bed.

In the case of mixing-cell model, packed bed is imagined to consist of a series of perfectly mixed tanks each on the scale of particle, but this model contains the parameters difficult to estimate.

It must be remarked that diffusion model or mixing-cell model is applicable for slight deviations from plug flow, but when the flow pattern of fluid deviates considerably from piston flow, then combined model or statistical model is chosen to represent fluid behavior.

The purpose of this investigation is to propose a mathematical model which can be satisfactorily characterize the mixing mechanism in packed bed, and to test the applicability of this model. In the work reported here, only singlephase flow is considered, and it is assumed that there is the probability distribution of fluid velocity over the cross-section of packed bed, and this distribution may be approximated by the normal distribution as proposed Townsend[1]. Existence of a distribution of fluid velocity gives rise to promote the longitudinal mixing in the same direction as the flow.

2. Mathematical model based on the probability-density function

It is assumed that a probability-density function of fluid velocity exists across any cross-section of packed bed and this function may be defined by following equation:

$$\int_{-\infty}^{\infty} f(U)\,dU = 1 \tag{1}$$

where $f(U)$ is probability-density function of fluid velocity, and $U \equiv u/\bar{u}$ is dimension-less velocity.

We now consider that packed bed consists of the bundles of such a tube of flow that fluid having the time of passage $\Delta\theta$ flows through it in plug flow at velocity u, and that the concentration of fluid at the outlet of each tube of flow becomes identical with the average concentration over cross-section of the bed.

From material balance on tracer component of fluid over a tube of flow, we can write the following equation:

$$\frac{\partial Y}{\partial \Theta} + \left(\frac{\bar{u}\theta_h}{L}\right) U \left(\frac{\partial Y}{\partial \zeta}\right) = 0 \tag{2}$$

where $Y = y/y_0$, $\Theta = \theta/\theta_h$ and $\zeta = z/L$ denote dimension-less concentration, time and distance, respectively.

Now a following relation holds

$$d\zeta/d\Theta = (\bar{u}\theta_h/L)\,U \tag{3}$$

From Eqs. (2) and (3), we have

$$dY/d\zeta = 0 \text{ and } dY/d\Theta = 0$$

Consequently, we obtain

$$Y(\Theta + \Delta\Theta,\ \zeta) = Y(\Theta,\ \zeta - \Delta\zeta). \tag{4}$$

The significance of this equation is agreed to the assumptions mentioned above.

Hence, the average concentration over cross-section $\bar{Y}$ at $\Theta = \Theta + \Delta\Theta$ and $\zeta = \zeta$ is obtained as follows:

$$\bar{Y}(\Theta + \Delta\Theta,\ \zeta) = \int_{-\infty}^{\infty} Y(\Theta + \Delta\Theta,\ \zeta) f(U)\,dU = \int_{-\infty}^{\infty} Y(\Theta,\ \zeta - \Delta\zeta) f(U)\,dU$$

$$\therefore \quad \bar{Y}(\Theta + \Delta\Theta,\ \zeta) = \int_{-\infty}^{\infty} Y\left(\Theta,\ \zeta - \frac{\bar{u}\theta_h \Delta\Theta}{L} U\right) f(U)\,dU \tag{5}$$

Eq. (5) is the mathematical representation taken into account of the density-distribution of fluid velocity. Calculations have been carried out to illustrate this model, we now consider the following example.

It is assumed that the probability-density function of fluid velocity conform to the Gaussian (normal) distribution as follows:

$$f(U) = \frac{1}{\sqrt{2\pi}\sigma}\exp\{-(U-1)^2/2\sigma^2\} \tag{6}$$

For simplicity, we suppose that the continuous function of normal distribution may be to approximate with the stepwise function.
For instance, putting

$$p_1 = f(U_1)\,\Delta U_1, \quad p_2 = f(U_2)\,\Delta U_2, \ldots, \quad p_k = f(U_k)\,\Delta U_k$$

And thereby Eq. (5) can be represented as follows:

$$\bar{Y} = \sum_i \{Y(\Theta, \zeta - \Delta\zeta) \cdot p_i\} \tag{7}$$

As the first approximation, dividing the abscissa U of normal distribution curve $f(U)$ vs. U into six intervals, and we construct a stepwise curve approximating a normal distribution curve. Setting the data for example as follows:

$$p_1 = p_6 = 0.0668, \quad p_2 = p_5 = 0.0919, \quad p_3 = p_4 = 0.3413$$

Thus calculated results when $\Delta\theta = 1$sec and $\bar{u} = 10$ cm/sec are illustrated in Fig. 1, and those when $\Delta\theta = 2$ sec and $\bar{u} = 10$ cm/sec are shown in Fig. 2. These figures indicate the variations of average concentration at each time-interval $\Delta\theta$ as a function of the upward distance from the bottom.

In order to compare our model proposed here with other models, we now reduce our model to the form of a partial differential equation.

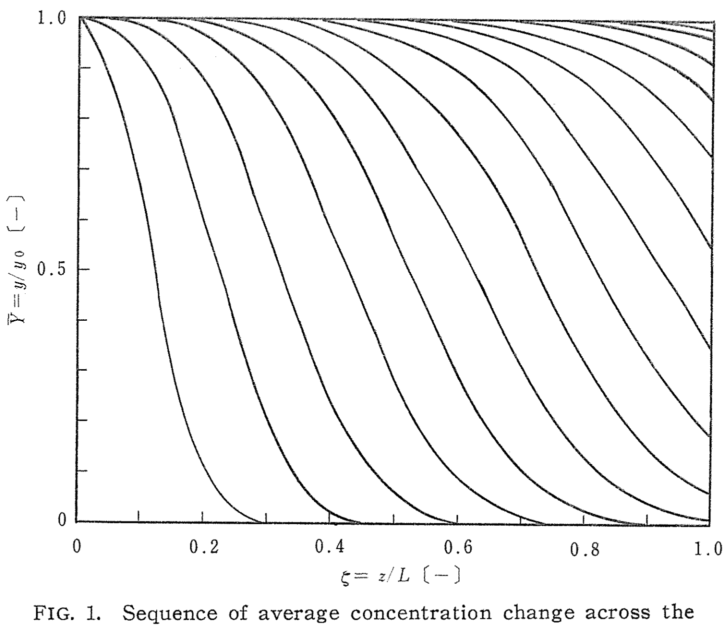

FIG. 1. Sequence of average concentration change across the cross-section of bed for each $\Delta\theta = 1$ sec.

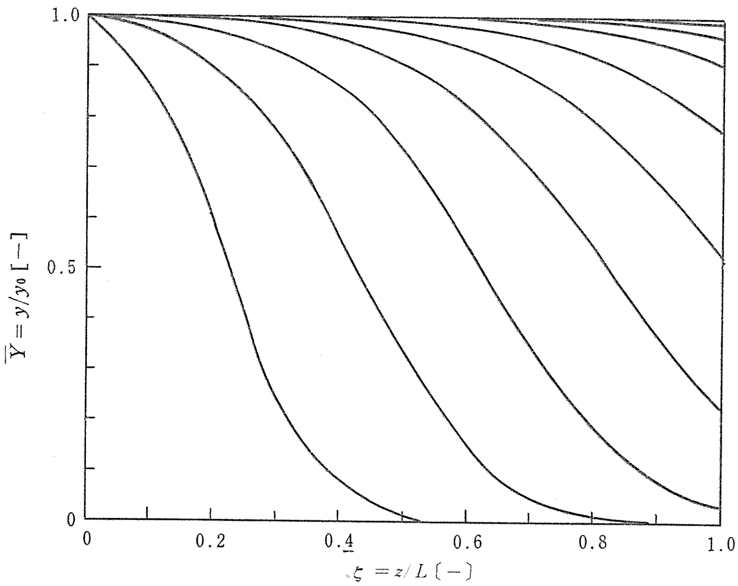

FIG. 2. Sequence of average concentration change across the cross-section of bed for each $\Delta\theta = 2$ sec.

Expansion of the both sides of Eq. (5) in the neighbourhood of (Θ, ζ) by means of Taylor's theorem gives

$$\overline{Y}(\Theta,\ \zeta) + \Delta\Theta\left(\frac{\partial\overline{Y}}{\partial\Theta}\right) + \frac{\Delta\Theta^2}{2!}\left(\frac{\partial^2\overline{Y}}{\partial\Theta^2}\right) + \cdots = Y(\Theta,\ \zeta) - \left(\frac{\bar{u}\theta_h\Delta\Theta}{L}\right)m\left(\frac{\partial Y}{\partial\zeta}\right) + \left(\frac{\bar{u}\theta_h\Delta\Theta}{L}\right)^2\frac{\sigma^2 + m^2}{2\,!}\left(\frac{\partial^2 Y}{\partial\zeta^2}\right) - \cdots \tag{8}$$

where m and σ^2 are dimension-less average velocity and variance from average velocity, respectively. And these parameters are defined as follows:

$$m = \int_{-\infty}^{\infty} Uf(U)\,dU$$

$$\sigma^2 = \int_{-\infty}^{\infty}(U - m)^2 f(U)\,dU$$

Now, from assumption above-mentioned, dimension-less concentration for fluid flowing out of a tube of flow can be written as follows:

$$Y(\Theta,\ \zeta) = \overline{Y}(\Theta,\ \zeta)$$

In Eq. (8), letting $\Delta\Theta$ be small sufficiently, then all the terms of higher order than third order on $\Delta\Theta$ may become negligible. Thus, replacing Θ which is involved in the third term of the left-hand side of Eq. (8) by ζ in terms of Eq. (3) and after re-arranging the expression we obtain a following equation:

$$\frac{\partial Y}{\partial\Theta} = -\left(\frac{\bar{u}\theta_h}{L}\right)m\frac{\partial Y}{\partial\zeta} + \frac{\sigma^2}{2}\left(\frac{\bar{u}^2\theta_h^2\Delta\Theta}{L^2}\right)\frac{\partial^2 Y}{\partial\zeta^2} \tag{9}$$

3. Comparison with other models

Diffusion model represented with the dimension-less form can be given as follows:

$$\frac{\partial Y}{\partial \Theta} = -\frac{\partial Y}{\partial \zeta} + \frac{E_z}{\bar{u}L}\frac{\partial^2 Y}{\partial \zeta^2} \tag{10}$$

Comparing Eqs. (9) and (10), w obtain

$$2\left(\frac{\bar{u}^2\theta_h^2 \Delta\Theta}{L^2}\right) = \frac{E_z}{\bar{u}L}$$

Since $\bar{u}\theta_h = L$ and $\theta_h \Delta\Theta = \Delta\theta$, therefore

$$E_z = \bar{u}^2 \Delta\theta\,(\sigma^2/2) \tag{11}$$

In Eq. (11), letting $\sigma \to \infty$, then $E_z \to \infty$, and as $\sigma \to 0$, $E_z \to 0$.

When the height of packed bed is sufficiently high, the relation between P_e in diffusion model and γ in mixing-cell model can be represented as follows[2) 3)].

$$P_e = \bar{u}D_p/E_z = 2/\gamma \tag{12}$$

where $\gamma = \lambda/D_p$, and λ is a height of packed bed equivalent to a perfectly mixed tank. From Eqs. (11) and (12), we have

$$\gamma = \bar{u}\Delta\theta/D_p\sigma^2 \tag{13}$$

and

$$\lambda = \bar{u}\Delta\theta/\sigma^2 \tag{14}$$

In the case of packed bed, when $R_e = D_p\bar{u}\rho/\mu$ becomes large, then γ approaches to 1.0, and so $E_z \to \bar{u}D_p/2$, $P_e \to 2$ and $\lambda \to D_p$. Thus $\sigma^2 \to \bar{u}\Delta\theta/D_p$ as $\gamma \to 1.0$.

Now, putting $\sigma^2 = 1$ in Eq. (14), then we have $\lambda = \bar{u}\Delta\theta$. This implies that a height of packed bed equivalent to a perfectly mixed tank in mixing-cell model becomes equal to a mean transfer distance of fluid in a time interval $\Delta\theta$ in our model.

Thus the correspondences of parameters between our model and diffusion and mixing-cell models have been given here clearly.

In order to compare the calculated results in terms of our model with those in diffusion model, F-diagram is illustrated in Fig. 3. In this figure, curves based on our model were obtained by using of the data of $\bar{Y}$ at $\zeta = z/L = 1.0$ in Figs. 1 and 2. From these curves, it is seen that the effect of mixing of fluid decreases with decrease in the value of $\Delta\theta$, and F-diagram approaches to the case of piston flow.

Among various mathematical models proposed in the past, Kunugita and Otake's model which is the extention of mixing-cell model resembles our model proposed here. They consider that a liquid flowing through packed bed consists of a number of minute portions of liquid, and that a portion of liquid stays at

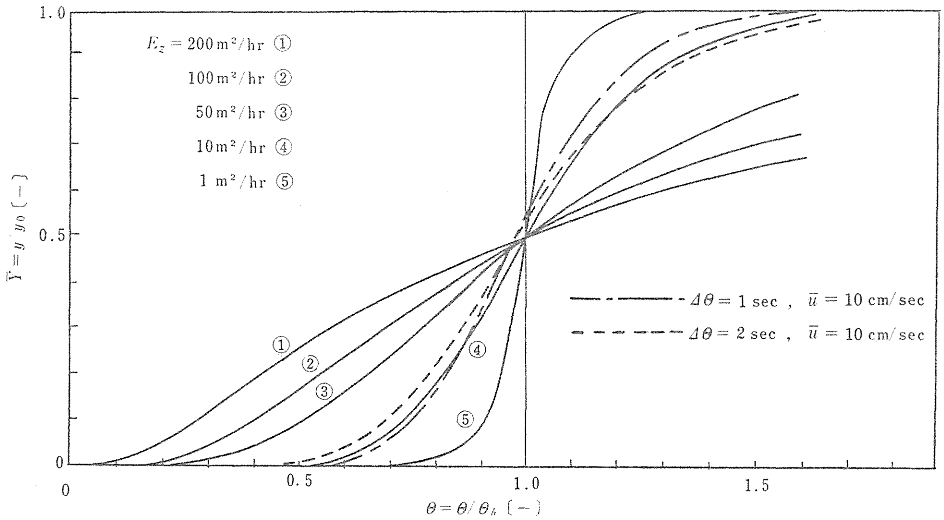

FIG. 3. Comparison of F-diagrams obtained by our model and those by diffusion model.

the same position and the remainders move in the positive or negative direction.

Letting p_γ and q_γ be the probability that liquid portion moves over the distance of γ-tanks in the positive and negative directions, respectively, and r be the probability that liquid portion stays at the same position, then the following relation holds.

$$\sum_\gamma p_\gamma + \sum_\gamma q_\gamma + r = 1 \tag{15}$$

Then, their model may be written as follows:

$$\bar{Y}(i, j+1) = \sum_\gamma p_\gamma Y(i-\gamma, j) + \sum_\gamma q_\gamma Y(i+\gamma, j) + rY(i, j) \tag{16}$$

Now, expanding the both sides of Eq. (16) in Taylor-series, taking into account up to the third term in each side, and re-arranging the expression, then we obtain

$$\frac{\partial Y}{\partial \Theta} = -\frac{m'}{\Delta\Theta}\frac{\partial Y}{\partial \zeta} + \frac{(\sigma')^2}{2\,\Delta\Theta}\frac{\partial^2 Y}{\partial \zeta^2} \tag{17}$$

where

$$m' = \sum_\gamma \{\gamma(p_\gamma + q_\gamma)\}\,\Delta\zeta,$$
$$(\sigma')^2 = \sum_\gamma \{\gamma^2(p_\gamma + q_\gamma)\}(\Delta\zeta)^2 - (m')^2$$

Hence, from comparison between Eq. (10) of diffusion model and Eq. (17), we obtain the following expressions:

$$E_z = (\sigma')^2/2\,\Delta\Theta$$
$$P_e = 2\,\bar{u}\Delta\Theta D_p/(\sigma')^2$$

Thus, expression for P_e reported by Kunugita and Otake has been modified as indicated above. In the case of Taylor-series expansion, they took only up to the second term into account and concluded that staying portion of liquid has no effect upon P_e.

Their model and our model are identical from the point of view setting the constant-time interval, but the distribution of fluid velocity is discrete in the former and it is continuous in the latter.

4. Application of model to moving-bed catalytic reactor

In order evaluate the practical value of our model, we now search for the distributions of concentration and temperatures in moving-bed catalytic reactor.

It is assumed here that:

(1) fluid flows through the bed according to the behavior which is given in our model, and solid particles flow down in plug flow;

(2) concentration and temperatures across any given cross-section of flow are even;

(3) reaction occurs on the surface of solid particle and is the n-th order irreversible reaction, and rate constant can be expressed by Arrhenius equation;

(4) physical properties of fluid and solid particle are constant.

If the above conditions are fulfilled, the process of longitudinal mixing and heat transfer in moving-bed catalytic reactor may be described by the following differential equations:

$$\frac{dY}{d\zeta} = -\frac{L}{2\bar{u}U} k y_0^{n-1} Y^n \left(\frac{1-\varepsilon}{\varepsilon}\right) \tag{18}$$

$$\frac{d\xi}{d\zeta} = \frac{L}{2\bar{u}U}\left(\frac{1-\varepsilon}{\rho_f C_f \varepsilon}\right)\left(\frac{6h_p}{\phi D_p}\right)\left(\frac{t_0}{T_0}\tau - \xi\right) \tag{19}$$

$$\frac{d\tau}{d\zeta} = -\frac{L}{2Vt_0}\left(\frac{\rho_f}{M\rho_s C_s}\right) Q k y_0^n Y^n + \frac{L}{2V\rho_s C_s}\left(\frac{6h_p}{\phi D_p}\right)\left(\frac{1}{1-\varepsilon}\right)\left(\tau - \frac{T_0}{t_0}\xi\right) \tag{20}$$

where $\xi = T/T_0$ and $\tau = t/t_0$ denote dimensionless temperatures of fluid and solid particle, respectively. Eqs. (18), (19) and (20) are derived from material balance on reactant and heat balances on fluid and on solid particle, respectively.

The boundary conditions are

$$Y = Y(\Theta,\ \zeta - \Delta\zeta),\ \xi = \xi(\Theta,\ \zeta - \Delta\zeta)\ \text{at}\ \zeta = \zeta - \Delta\zeta,$$

$$\tau = \tau(\Theta,\ \zeta + \Delta\zeta)\ \text{at}\ \zeta = \zeta + \Delta\zeta$$

These equations cannot be solved analytically. Hence, in order to obtain approximate solutions, setting k=constant in Eq. (18), $\tau - (T_0/t_0)\xi$=constant in Eqs. (19) and (20) and kY^n=constant in Eq. (20) for the range of sufficiently small $\Delta\zeta$ or $\Delta\Theta$.

Thus the solution of Eq. (18) for $n \neq 1$:

$$Y(\Theta + \Delta\Theta,\ \zeta) = [\{(n-1)Lky_0^{n-1}(1-\varepsilon)/(2\bar{u}U\varepsilon)\}\Delta\zeta + \{Y(\Theta,\ \zeta - \Delta\zeta)\}^{n-1}]^{1/(n-1)} \tag{21}$$

For $n = 1$:

$$Y(\Theta+\Delta\Theta,\ \zeta)=Y(\Theta,\ \zeta-\Delta\zeta)\exp\left\{-\frac{kL(1-\varepsilon)\Delta\zeta}{\bar{u}U\varepsilon}\right\} \tag{22}$$

The solution of Eq. (19):

$$\xi(\Theta+\Delta\Theta,\ \zeta)=\frac{L}{2uU}\left(\frac{1}{\rho_f C_f}\right)\frac{6h_p}{\phi D_p}\left(\frac{1-\varepsilon}{\varepsilon}\right)\left(\frac{t_0}{T_0}\tau-\xi\right)\Delta\zeta+\xi(\Theta,\ \zeta-\Delta\zeta) \tag{23}$$

The solution of Eq. (20):

$$\tau(\Theta+\Delta\Theta,\ \zeta)=\tau(\Theta,\ \zeta+\Delta\zeta)-\frac{L\rho_f Qky_0^n}{2(1-\varepsilon)Vt_0M\rho_sC_s}Y^n+\frac{6h_pL}{2V\phi D_p\rho_sC_s}\left(\tau-\frac{T_0}{t_0}\xi\right)\Delta\zeta\frac{1}{1-\varepsilon} \tag{24}$$

Now, average concentration and average temperatures of fluid and solid particles across the cross-section of moving-bed can be written as follows:

$$\bar{Y}(\Theta+\Delta\Theta,\ \zeta)=\int_{-\infty}^{\infty}Y(\Theta+\Delta\Theta,\ \zeta)f(U)dU \tag{25}$$

$$\bar{\xi}(\Theta+\Delta\Theta,\ \zeta)=\int_{-\infty}^{\infty}\xi(\Theta+\Delta\Theta,\ \zeta)f(U)dU \tag{26}$$

$$\bar{\tau}(\Theta+\Delta\Theta,\ \zeta)=\int_{-\infty}^{\infty}\tau(\Theta+\Delta\Theta,\ \zeta)f'(U)dU \tag{27}$$

Therefore, $\bar{Y}$, $\bar{\xi}$ and $\bar{\tau}$ may be obtained by means of Eqs. (21)-(27). Namely, we now substitute Eqs. (21) or (22), (23) and (24) into the right-hand sides of Eqs. (25), (26) and (27), respectively. And, repeating the calculations based on these equations at the beginning and end of the time interval, then longitudinal distributions of concentration and temperatures may be obtained.

Improved results may be effected by decreasing the size of the interval. In particular, when $|t-T|\ \Delta\theta$ has a large value, it is necessary to take sufficiently small size of the interval.

5. Conclusion

In order to determine the mixing characteristics of fluid in packed bed, mathematical model basing on the probability-density distribution of fluid velocity has been proposed, and it has been compared with other models.

From the comparison with diffusion model at the condition of limitation $\Delta\theta\to 0$, the relations between our model and diffusion or mixing-cell models have been indicated.

When we inquire into the transient response, a troublesome trial and error calculus is needed in diffusion model, but in our model there is no need to lean on such a calculus.

Nomenclature

C_f	: specific heat of fluid	[kcal/kgmol・°C]
C_s	: specific heat of solid particle	[kcal/kg・°C]
D_p	: particle diameter	[m]

E_z	:	longitudinal dispersion coefficient	[m²/hr]
G_f	=	$u_0 \rho_f$	[kgmol/m² • hr]
G_s	=	$V\rho_s$	[kg/m² • hr]
h_p	:	particle-to-fluid heat transfer coefficient	[kcal/m² • hr • °C]
k	:	reaction rate constant	[1/hr]
$\bar{k}$	:	reaction rate constant	[m³/kg(cat.) • hr]
L	:	bed height	[m]
M	=	WR/N	[−]
N	=	$6\,h_p(1-\varepsilon)L/\phi D_p G_f C_f$	[−]
P_e	=	Peclet number	[−]
Q	:	heat generated by reaction	[kcal/kgmol]
R	=	$\bar{k}(1-\varepsilon)\rho_s L/u_0$	[−]
T	:	temperature of fluid	[°C]
t	:	temperature of solid particle	[°C]
u	:	linear velocity of fluid in bed	[m/hr]
$\bar{u}$	:	mean velocity of fluid in bed	[m/hr]
u_0	:	superficial velocity of fluid	[m/hr]
V	:	mean velocity of solid particle	[m/hr]
W	=	$G_s C_s/G_f C_f$	[−]
y	:	molar fraction	[−]
y_0	:	y at $z=0$	[−]
z	:	axial distance from bottom of bed	[m]
ε	:	average fractional void in bed	[−]
θ	:	time	[hr]
θ_h	:	mean holding time	[hr]
ρ_f	:	density of fluid	[kgmol/m³]
ρ_s	:	pellet density of solid particle	[kg/m³]
ϕ	:	shape factor of solid particle	[−]

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