

Supplementary Material



Heat transfer analysis of water phase flowing in a PTFE tube upon heating

Supplementary Scheme 1. A model of water phase flowing in a PTFE tube immersed in an oil bath.

Supplementary Scheme 1 shows a model of water phase flowing in a PTFE tube immersed in an oil bath at 70 °C. Here, we calculate how a temperature of the inner fluid (water) increases from 20 °C when the water passes through a PTFE tube with a fixed length of L m at 70 °C.

Since the amount of heat obtained by the water flowing in a circular tube is equal to the amount of heat passing through a thermal boundary layer per unit time, the heat balance equation for outlet temperature T is expressed by equation (1).

$$C_p W (T - T_c) = U A \frac{\Delta T_1 - \Delta T_2}{\ln(\Delta T_1 / \Delta T_2)} = U A \frac{(T_H - T_c) - (T_H - T)}{\ln((T_H - T_c) / (T_H - T))}$$
(1)

where C_p [J kg⁻¹ K⁻¹] is the heat capacity at a constant pressure, W [kg s⁻¹] is the mass flow rate, T [°C] is the outlet temperature at a fixed tube length of L [m], T_c [°C] is the temperature of a cold fluid (water), T_H [°C] is the temperature of a hot fluid (oil bath), U [W m⁻² K⁻¹] is the overall heat transfer coefficient, A is the heat transfer surface area.

The overall heat transfer resistance $(\frac{1}{UA})$ is defined as the sum of thermal boundary resistances in each layer as shown in equation (2).

$$\frac{1}{UA} = \frac{1}{h_H A_{out}} + \frac{l}{\lambda_{wall} A_{lm}} + \frac{1}{h_c A_{in}}$$
(2)

where $h [W m^{-2} K^{-1}]$ is the heat transfer coefficient at oil bath (H) and water phase (C), $\lambda_{wall} [W m^{-1} K^{-1}]$ is the thermal conductivity of a wall (PTFE tube), $A_{out} [m^2]$ is the outer surface area of the PTFE tube, $A_{in} [m^2]$ is the inner surface area of the PTFE tube, and $A_{lm} [m^2]$ is the logarithmic mean of the inner and the outer area of the PTFE tube.

The first term on the right hand of the equation (2) is the heat transfer resistance from a hot fluid (oil bath) to a solid surface (PTFE) *via* convection heat transfer. In our system, this term can be negligible because the temperature of the oil bath is kept constant at 70 °C.

The second term on the right hand of the equation (2) is the heat transfer resistance in a sold wall (PTFE) *via* thermal transfer. We calculated the second term using characteristic values and the results are shown in **Supplementary Table 1**.

 $A_{in} = 0.001 \text{ [m] (I.D. of tube)} \times \pi \times 0.1 \text{ [m] (Length of tube under heating)} = 3.14 \times 10^{-4} \text{ [m^2]}$ $A_{out} = 0.00159 \text{ [m] (O.D. of tube)} \times \pi \times 0.1 \text{ [m] (Length of tube under heating)} = 5.0 \times 10^{-4} \text{ [m^2]}$ $A_{lm} = \frac{A_{out} - A_{in}}{ln \frac{A_{out}}{A_{in}}} = 4.0 \times 10^{-4} \text{ [m^2]}$

$\lambda_{wall} \; [\mathrm{W} \; \mathrm{m}^{-1} \; \mathrm{K}^{-1}]$	<i>l</i> [m]	$A_{in} [\mathrm{m}^2]$	$A_{out} [m^2]$	A_{lm} [m ²]	$l/\lambda_{wall}A_{lm}$ [K W ⁻¹]
0.23	0.59×10^{-3}	3.14 × 10 ⁻⁴	5.0×10^{-4}	4.0×10^{-4}	6.4

Supplementary Table 1. Characteristic values to calculate overall heat transfer resistance.

The third term on the right hand of the equation (2) is the heat resistance from a solid surface (PTFE) to a cold fluid (water) *via* heat transfer. Water a flow in the PTFE tube can be described by the Reynolds number (Re [-]) as shown in equation (3)

$$Re = \frac{\rho \bar{u} D}{\mu} \tag{3}$$

where ρ [kg m⁻³] and μ [Pa s] are density and viscosity of water at 45 °C (which is a mean temperature between 20 and 70 °C), \bar{u} [m s⁻¹] is a linear flow rate of the fluid, and D [m] is the inner diameter of the PTFE tube. Calculation result of *Re* is shown in **Supplementary Table 2**.

Cross section of the channel $[m^2] = (\frac{0.001 \, [m]}{2})^2 \times \pi = 7.85 \times 10^{-7} \, [m^2]$ $\bar{u} \, [m \, s^{-1}] = \frac{184.6 \, [\mu L \, min^{-1}] \times 10^{-9}}{60 \, [s]} \, [m^3 \, s^{-1}] / \, 7.85 \times 10^{-7} \, [m^2] = 3.92 \times 10^{-3} \, [m \, s^{-1}]$ $Re = \frac{\rho \bar{u} D}{\mu} = \frac{990.22 \, [kg \, m^{-3}] \times 3.92 \times 10^{-3} [m \, s^{-1}] \times 0.001 \, [m]}{0.000598 \, [Pa \, s]} = 6.49 \, [-]$

	Flow rate		Linear flow rate in a tube	
Dispersed phase	Continuous phase	Total	\bar{u}	Re [-]
$[\mu L min^{-1}]$	$[\mu L min^{-1}]$	$[\mu L min^{-1}]$	$[m \ s^{-1}]$	
92.3	92.3	184.6	3.92×10^{-3}	6.5

Supplementary Table 2. Linear flow rate and Reynolds number in the slug flow system.

The Nusselt number (Nu) is the ratio of convective to conductive heat transfer across a boundary and is described by equation (4).

$$Nu = \frac{hD}{\lambda_{fluid}} \tag{4}$$

where λ_{fluid} [W m⁻¹ K⁻¹] is the thermal conductivity of the fluid (water). In this system, *Re* is relatively low, which indicates that a flow in a microchannel is dominated by laminar flow. Despite of the laminar flow condition, natural convection in a microchannel can be negligible due to relatively small temperature difference between microchannel wall and water phase in the channel. Then, *Nu* is expressed by equation (5).

$$Nu = 1.86 \left(RePr \frac{D}{L} \right)^{1/3} \left(\frac{\mu}{\mu_w} \right)^{0.14}$$
(5)

where Pr [-] is the Prandtl number, L [m] is the length of a tube, μ [Pa s] is the viscosity of fluid, μ_w is the viscosity of a fluid at the mean wall temperature.

From equation (4) and (5), the heat transfer coefficient (h) is described by equation (6).

$$h = 1.86 \left(RePr \frac{D}{L} \right)^{1/3} \left(\frac{\mu}{\mu_w} \right)^{0.14} \left(\frac{\lambda}{D} \right)$$
(6)

Next, the Prandtl number (Pr [-]) is a dimensionless number, defined as the ratio of momentum diffusivity to thermal diffusivity. Pr is described by equation (7).

$$Pr = \frac{C_p \mu}{\lambda_{fluid}} \tag{7}$$

Then, we calculated the heat transfer coefficient (*h*) using characteristic values in **Supplementary** Table 3.

$$Pr = \frac{C_p \mu}{\lambda_{fluid}} = \frac{4179 \left[J \ kg^{-1}T^{-1} \right] \times 0.000598 \left[Pa \ s \right]}{0.637 \left[W \ m^{-1} \ K^{-1} \right]} = 3.92$$

$$Nu = 1.86 \left(RePr \frac{D}{L} \right)^{1/3} \left(\frac{\mu}{\mu_w} \right)^{0.14}$$

= 1.86 × (3.90 [-] × 3.92 [-] × $\frac{0.001 [m]}{0.1 [m]}$)^{1/3} × ($\frac{0.000598 [Pa s]}{0.000404 [Pa s]}$)^{0.14} = 1.25

$$h_C = Nu\left(\frac{\lambda}{D}\right) = 1.25 \left[-\right] \times \left(\frac{0.637 \left[W \ m^{-1} \ K^{-1}\right]}{0.001 \ [m]}\right) = 793 \left[W \ m^{-2} \ K^{-1}\right]$$

Supplementary Table 3. Characteristic values to calculate the heat transfer coefficient $h_{\rm C}$.

Re [-]	Pr [-]	Nu [-]	$h_{C} [{ m W} { m m}^{-2} { m K}^{-1}]$	A_{in} [m ²]	$1/h_{\mathcal{C}}A_{in}$ [K W ⁻¹]
6.5	3.9	1.3	793	3.14× 10 ⁻⁴	4.0

Using these parameters, we calculated the overall heat resistance and the overall heat transfer

coefficient as shown in Supplementary Table 4.

$$\frac{1}{UA} = \frac{1}{h_H A_{out}} + \frac{l}{\lambda_{wall} A_{lm}} + \frac{1}{h_c A_{in}} = 4.0 + 6.4 = 10.4 \ [K \ W^{-1}]$$

Supplementary Table 4. Overall heat transfer coefficient calculated using the model.

Re [-]	$l/\lambda_{wall}A_{lm}$ [K W ⁻¹]	$1/h_{C}A_{in}$ [K W ⁻¹]	1/UA [K W ⁻¹]	<i>UA</i> [W K ⁻¹]
6.5	4.0	6.4	10.4	0.09587

Finally, we calculated the outlet temperature (T) using the equation (1).

From the equation (1),

 $Q = UAT_{lm}$

Here,
$$T_{lm} = \frac{\Delta T_1 - \Delta T_2}{\ln (\Delta T_1 / \Delta T_2)} = \frac{(T_H - T_c) - (T_H - T)}{\ln ((T_H - T_c) / (T_H - T))} = \frac{(70 - 20) - (70 - T)}{\ln ((70 - 20) / (70 - T))} = \frac{T - 20}{\ln (50 / (70 - T))}$$

Then,

 $Q = UAT_{lm}$ $= UA\frac{T-20}{\ln\left(\frac{50}{70-T}\right)} \qquad (8)$

 $Q = C_p W (T - 20)$ (9)

From the equation (8) and (9),

$$C_p W (T-20) = UA \frac{T-20}{\ln\left(\frac{50}{70-T}\right)}$$
$$\ln\left(\frac{50}{70-T}\right) = \frac{UA}{C_p W}$$
$$T = 70 - \frac{50}{\exp\left(\frac{UA}{C_p W}\right)}$$
$$T = 69.9889 \approx 70 [^{\circ}C]$$

By changing the length of the PTFE tube, we analyzed the change in the temperature of water slugs flowing in the microchannel after entering the heating zone as shown below. This result clearly suggests that the temperature of water slugs after entering the heating zone reaches at 70 °C in 18 seconds.



Supplementary Figure 1. Change in the temperature of water slugs after entering a heating zone as a function of time.